

High Sensitive Biosensors with Slot and Stack-Type Structure using Silicon Nitride Waveguides

Yoshiteru Amemiya¹, Anna Hirowatari¹, Tomoya Taniguchi¹, Takeshi Ikeda^{1,2}, Masataka Fukuyama¹, Akio Kuroda^{1,2}, and Shin Yokoyama^{1,*}

¹Research Institute for Nanodevice and Bio Systems, Hiroshima University

²Department of Molecular Biotechnology, Graduate School of Advanced Sciences of Matter, Hiroshima University
1-4-2 Kagamiyama, Higashihiroshima, Hiroshima 739-8527, Japan

Phone: +81-82-424-6265, FAX: +81-82-424-3499, E-mail: yokoyama-shin@hiroshima-u.ac.jp

1. Introduction

Early detection and early treatment of diseases are very important especially for the aging society. Immunoassay is a detection method of the diagnosis of the diseases and Enzyme-Linked Immuno-Sorbent Assay (ELISA) is widely used, where antigen or antibody is labeled with enzyme or fluorescent materials. For simple and easy detection, we have investigated a biosensor chip using silicon-binding protein (designated as Si-tag) and ring resonator as shown in Fig. 1 [1, 2]. The advantages of Si-tag are high sensitivity and the ability of label-free detection, because Si-tag directly binds to silicon oxide (SiO₂) or silicon nitride (SiN) surface in an oriented manner as an anchoring molecule to immobilize receptor (Fig. 1). The advantages of ring resonator are easily integration of small size devices and the fabrication of multiple detection system using the Si semiconductor technologies.

The previous detection sensitivity of our method was an order of 10⁻⁷ g/ml [2] and should be increased by about two orders more, because the sensitivity of the order 10⁻⁹ g/ml is generally required for the diagnosis of diseases. For this purpose, we evaluate SiN slot ring resonator, and propose the stack-type slot ring resonator for further high sensitivity. Because the optical electric-field is concentrated at sensing part in slot ring, the detection sensitivity becomes higher than normal ring resonator. The usage of SiN has several advantages, easily integration of Si p/n photodiode detector, low temperature dependence of refractive index, and so on.

2. Experimental

The SiN slot ring biosensor is fabricated by the two-step chemical vapor deposition method as shown in Fig. 2. This method is used to prevent cracks of SiN caused by the high internal stress of the film > 0.3 μm. The 0.25-μm-thick SiN was firstly deposited and etched them, and the rest of 0.25 μm was deposited and etched once more. The scanning-electron-microscope (SEM) image of the fabricated device is shown in Fig. 3. The slot width, waveguide width and height are 0.1-0.2, 0.5-0.6 and 0.5 μm, respectively.

A biomaterial can be sensed by measuring a change in optical output intensity through the ring resonator, where the refractive index change and resonance wavelength shift are caused by the biomaterial adsorbing on the ring resonator by dropping the biomaterial solution. For the reducing optical absorption by solution [3], the laser with wavelength 1300 nm is used. And output optical intensity is detected by InGaAs detector.

Resonance wavelength shifts are simulated using the finite element method (FemSIM "RSoft Design Group) and

can be compared with measured results. Here, refractive index of the solution, the SiN and the biomaterials (Si-tag, biotin, and so on) are assumed to be 1.33, 2.00 and same as SiO₂ of 1.45, respectively. The thickness of Si-tag and detected biomaterial are set to be 10 and 5 nm, respectively.

3. Results and Discussion

The temperature dependence of the resonance characteristics of the SiN slot ring resonator is shown by Fig. 4. The resonance wavelength shift $\Delta\lambda_{\text{res}}$ is plotted as a function of the temperature change ΔT in inset, which results in the temperature coefficient of 0.006 nm/°C [4] while that of the Si ring resonator without slot is one order high, 0.06 nm/°C [2]. This result leads lower thermal noise and it is the most useful advantage of SiN ring resonator for biosensor.

The performance of fabricated biosensor is evaluated using biotin and streptavidin reaction which are well known reaction. The Si-tagged biotins were adsorbed on the surface of ring. Then, the streptavidins were added in the sample solution (see Fig. 5). In Fig. 6, the resonance wavelength shift is plotted as a function of the concentration of the streptavidin solution. While the previous Si ring biosensor detects 10⁻⁷ g/ml streptavidin [2], the SiN slot ring biosensor has one order better sensitivity of 10⁻⁸ g/ml [4].

For the practical sensitivity of 10⁻⁹ g/ml, we propose stack-type slot ring resonator as shown in Fig. 7. The stack-type slots with expected height are easily fabricated by Si₃N₄/SiO₂ multi-layers deposition and HF etching of these SiO₂ layers. This easy control of slot height is the advantage of stack-type slot compared with lateral-type slot. Figure 8 shows the slot height h_s dependence of the simulated refractive index change Δn between with and without detected biomaterial layer as a parameter of the number of slots, k . Here, total slot waveguide height h_T is $(1.0+h_s)$ μm for each h_s . The Δn at the design of fabricated biosensor is about 4 times of Si ring without slot. The $\Delta\lambda_{\text{res}}$ is theoretically proportional to Δn , but the measured ratio of $\Delta\lambda_{\text{res}}$ between Si ring without slot and SiN slot ring is different from 4 at high concentration (see Fig. 6). One of the reasons for this difference is the simple simulation setup where detected biomaterial is uniformly adsorbed at one layer of 5 nm. Practically, the adsorption is complex and separates from simple simulation setup. Figure 9 shows slot waveguide height h_w , which is the height of one SiN layer, dependence of the refractive index change with 4 slots where the slot height h_s is fixed at 0.04 μm. Optimal value of h_w or h_T is about 0.12 or 0.8 μm and Δn becomes about 4.5 times of the fabricated biosensor with one slot. Because

$\Delta\lambda_{res}$ becomes also 4.5 times of the fabricated biosensor, detection sensitivity is improved more than one order of magnitude high (e.g., $\Delta\lambda_{res}$ becomes about 0.4 nm at 10^{-8} g/ml, which is easily detected, see Fig. 6). The practical sensitivity of 10^{-9} g/ml is expected to be achieved.

4. Conclusion

In this work, we have evaluated SiN slot ring resonator and the one order improvement of the detection sensitivity compared with the previous biosensor using the simulation and measurement. For the further improvement, we proposed stack-type slot ring resonator. In the simulation, about 4.5 times of resonance wavelength shift is estimated and the achievement of practical sensitivity of 10^{-9} g/ml is expected.

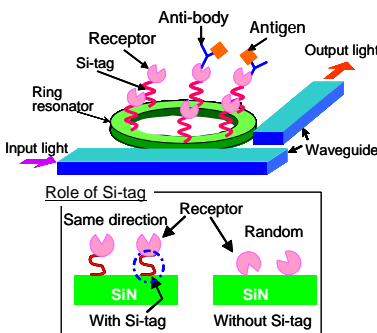


Figure 1. Basic concept of ring resonator biosensor. Receptors are aligned in same direction due to Si-tag.

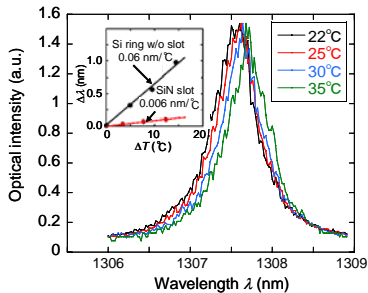


Figure 4. Temperature dependence of the SiN slot ring. Inset is resonance wavelength shift of Si ring without slot and SiN slot ring versus temperature change.

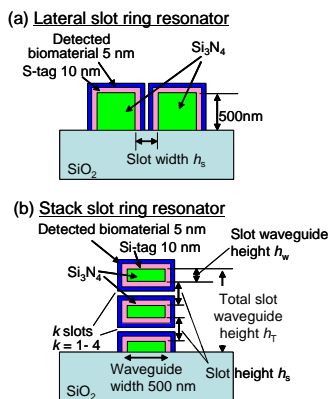


Figure 7. Schematic structure for the simulation (a) lateral-type and (b) stack-type slot ring resonator.

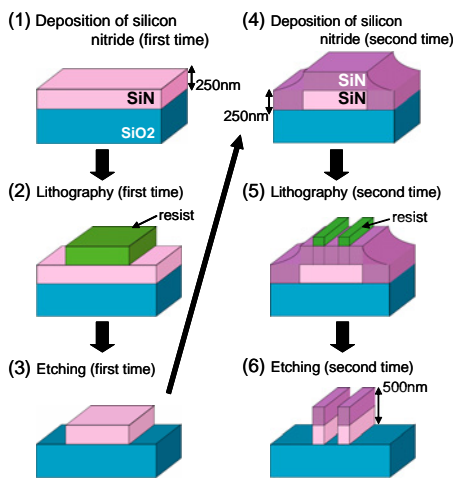


Figure 2. The processes of the two-step chemical vapor deposition method.

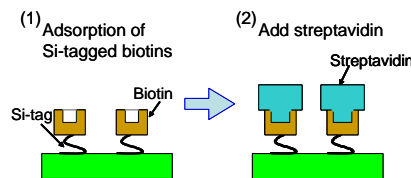


Figure 5. Experimental procedure for detection of the reaction of biotin with streptavidin.

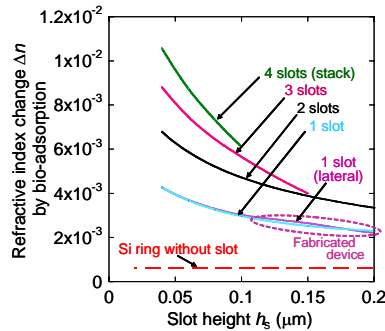


Figure 8. Simulated slot height dependence of refractive index change where total slot waveguide height h_T is $(1.0+h_s)$ μ m. Lateral-type slot ring with one slot is almost same as stack-type with one slot.

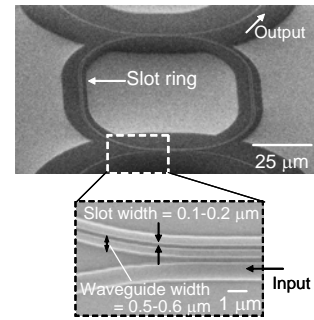


Figure 3. Bird's eye view of the SEM image of SiN slot ring resonator and expanded view of the gap region.

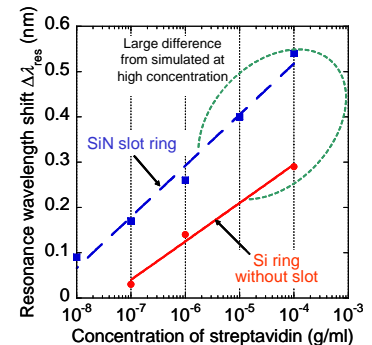


Figure 6. Comparing of SiN slot ring and Si ring without slot for the detection of biotin-streptavidin reaction.

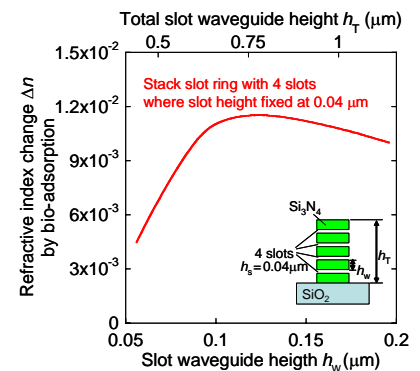


Figure 9. Simulated slot waveguide height dependence of refractive index change of stack-type slot with 4 slots where the slot height h_s is 0.04 μ m.

Acknowledgements

This work was supported in part by a Grant-in-Aid for Young Scientists (B) (No. 24760281) from the Ministry of Education, Culture, Sports, Science and Technology of Japan, and Industrial Technology Research Grant Program in 2009 from New Energy and Industrial Technology Development Organization of Japan (NEDO).

References

- [1] S. Yamatogi *et al.*, Jpn. J. Appl. Phys. **48** (2009) 04C188.
- [2] M. Fukuyama *et al.*, Jpn. J. Appl. Phys. **50** (2011) 04D107.
- [3] K. F. Palmer *et al.*, J. Opt. Soc. Am. **64** (1974) 1107.
- [4] A. Hirowatari *et al.*, Proc. of SPIE Photonics Europe 2012, **8431** (2012) 63.