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Effect on Sensitivity of Packaging Materials Used in a Microelectrode Sensor

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マイクロ電極センサの応答性に対するパッケージング材料の影響

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概要 筆者らはこれまでに，チップ上に実装した櫛形電極に電解質溶液(NaCl)を滴下し上部から光を照射すると，光強度，ならびにイオン濃度に応じて電極間の抵抗値が変化することを報告している。本研究では基板上に構築したセンサのパッケージング材料について検討した。パッケージングは下基板側にセンサを構築し，フィルム状あるいは板状の上基板で封止した。上下基板の間にはチャンバーを設け，上基板側の小孔からNaCl溶液を注入した。上基板を種々の材料に変え，光照射強度一定下でNaCl濃度に対する感度を調べた。テフロンは検出感度に影響を及ぼさなかった。その他の材料は，特に低濃度のNaCl溶液の場合に検出感度を大きく低下させた。この影響は，上基板の膜厚に依存した。またNaCl濃度を一定にした状態で，光照射強度に対する感度を調べた。この場合，テフロン，ポリプロピレン，ポリクロロビニリデンは光検出感度に影響しなかった。この影響も上基板の膜厚に依存した。基板に実装したマイクロ電極センサの検出感度は，パッケージングの材質と膜厚に依存することがわかった。

Key Words: *Sensor, Chemical Sensor, Photo Sensor, MEMS, Packaging Material*

1. Introduction

We previously developed a microsensor consisting of a pair of electrodes (2 mm ϕ) capable of measuring ion concentrations in NaCl, amino acids or nucleic acid solutions (1 nl~1 μl)[1]. Of minimal size and with a simple structure it has an enormous advantage in that it can easily be mounted on a flat chip or surface. The microsensor can detect changes in voltage or conductivity when the ion concentration is altered in a drop of NaCl, amino acid or nucleic acid solution on the sensor surface. The electrical resistance of the optimized microsensor is capable of detecting NaCl concentrations within the range of 1.0×10^{-4}~1.0 mol l^{-1}.

We have also reported that the microsensor is responsive to changes in light intensity when an electrolyte solution is applied to the microelectrode surface during white light irradiation, with the optical sensitivity observed being similar to that displayed by certain CdS semiconductor sensors[2].

In this study, we aimed to develop a microsensor chip that can detect both chemical concentration and light intensity. We now report the effects of the packaging materials on the limits of detection of NaCl concentration and optical sensitivity irradiated on the microelectrode.

2. Experiments

An experimental measurement system for determining concentration and optical sensitivity of a microsensor is shown in Figure 1(a). The microelectrode [No. 5] fabricated on the bottom board [No. 6] was connected to an analog to digital converter [No. 9] via an interface board [No. 8]. An upper transparent board [No. 2] was mounted on the bottom board and light was irradiated onto the upper board by a white LED [No. 1]. Figure 1(b) shows the top view of the upper transparent board. A photolithography method was used to pattern microelectrodes on an epoxy resin chip as the bottom board (25×25 mm, and 1 mm thick). Rectangular-shaped Cu electrodes were fabricated at 0.2 mm (width), 3.5 mm (length), and inter-electrode spacing (0.7 mm). The bottom board also had a micro well [No. 7] connected to a micro channel [No. 10]. The upper transparent board had two pin-holes that functioned as the entrance [No. 3] and exit [No. 4] for liquids. These micro structures were fabricated using an NC lathe method. The micro chamber [No. 11] was filled by injecting NaCl solution at the entrance hole.

The electrical resistance of the microsensor was measured after the NaCl solution (10^{-4}~1 mol l^{-1}) was ap-

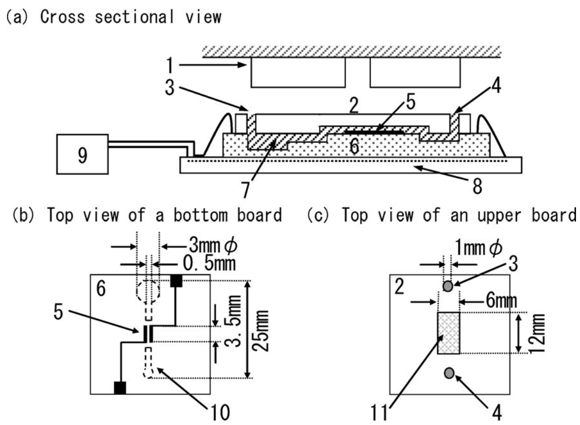

Fig. 1 (a) Schematic diagram of the measurement system for determining optical and concentration sensitivity of a microsensor. (b) Top view of the bottom board. (c) Top view of the upper transparent board

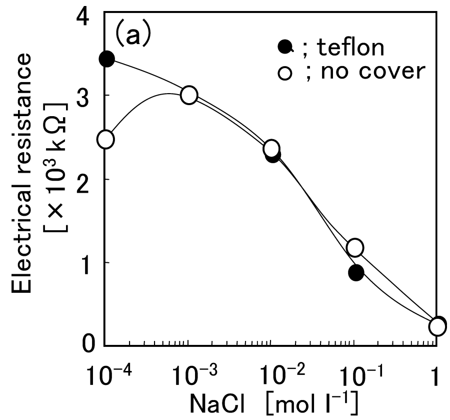

Fig. 2 Relationship between the electrical resistance of the microsensor and the concentration of the NaCl solution (a)

Upper board material: teflon film and no cover (reference).

plied to the chamber under white light irradiation of 0～15000 Lux emitted from an LED.

3. Results and Discussion

Figure 2 shows the electrical resistance of microelectrode dependence on NaCl concentration, when irradiated by white light at 500 Lux. When a teflon (TF) film (80 μm thick) was used as the upper transparent board, the microsensor exhibited high responsiveness within an NaCl concentration of 10^{-4}～$1\,\mathrm{mol\,l^{-1}}$. Electrical resistance showed a marked increase with decreased NaCl concentration. A microsensor that possessed no upper board (no cover) exhibited responsiveness at NaCl concentrations of 10^{-3}～$1\,\mathrm{mol\,l^{-1}}$, although the electrical resistance decreased in a $10^{-4}\,\mathrm{mol\,l^{-1}}$ NaCl solution.

Figure 3 shows the electrical resistance of microelectrode dependence on NaCl concentration at an irradiated light intensity of 500 Lux. In this case, polypropylene (PP) and PET films, 250 μm and 350 μm thick, respectively, were used as the upper transparent board. The microsensor covered TF film showed a linear electrical resistance with respect to NaCl concentration, whereas the electro resistance of the microelectrode covered with PP or PET film showed only a slight correlation within a range of 10^{-2}～$1\,\mathrm{mol\,l^{-1}}$. Interestingly, electrical resistance was observed to decrease under $10^{-3}\,\mathrm{mol\,l^{-1}}$ of NaCl concentration.

Figure 4 shows the electrical resistance of the microelectrode's dependence on NaCl concentration. In this case, polyimide (PI) (40 μm), polychlorovinylidene (PCV) (10 μm), and silicon rubber (SI) (500 μm) films, and glass (150 μm) were used for the upper transparent

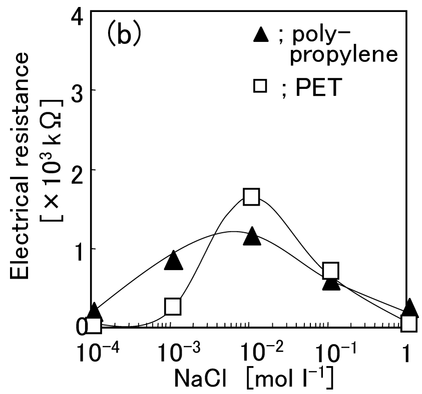

Fig. 3 Relationship between the electrical resistance of the microsensor and the concentration of the NaCl solution (b)

Upper board material: polypropylene and PET films.

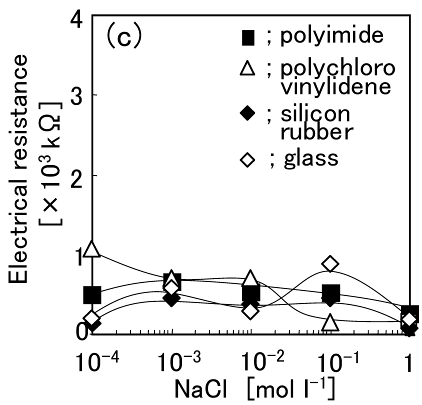

Fig. 4 Relationship between the electrical resistance of the microsensor and NaCl concentration (c)

Upper board material: polyimide, polychlorovinylidene, and silicon rubber films, and glass.

board. Experimental conditions were the same as those outlined in Figures 2 and 3. Microsensors covered with those materials showed linear electrical resistances with respect to the NaCl concentration.

Figure 5 shows the relationship between the electrical resistance of the microsensor and the thickness of various kinds of upper board materials under an irradiated light intensity of 500 Lux. When the concentration of NaCl in a chamber was low [Figure 5(a); 10^{-4} mol l^{-1}], the electrical resistance of the sensor was markedly reduced with increasing thickness of the upper board. When thickness of the upper board was above 40 μm, the electrical resistance showed a constant value, although TF films did not show this tendency. When the concentration of NaCl in the chamber was high [Figure 5(b); 1 mol l^{-1}], the electrical resistance of the sensor was lower and of a constant value; further, it was not related to the thickness or the material of the upper board.

It is widely known that an ion layer, like an electric double layer, is formed at the surface of solids and liquids[2]. Concentration distributions of Na^+ or Cl^- are higher at the surface of the sensor electrode[2]. It is also estimated that the distribution of ions is higher at the inside surface of the liquid and the upper board, and that the area of the electric double layer is increased. This would be the reason that electric conductivity markedly increases or electrical resistance markedly reduces.

Figure 6 shows the electrical resistance from the microelectrode's dependence on light irradiation. In each case, 10^{-4} mol l^{-1} of NaCl solution was filled into the micro chamber. The electrical resistance of microelectrodes covered with PP or TF films showed good correlations with light irradiation. Electrical resistance decreased with increases of the light intensity. The same trend was observed when the microelectrode was covered with PCV film or when there was no cover, although the electrical resistance was lower than that with PP or TF film covers. In contrast, the microelectrodes covered with PI, PET, or SI films, or glass showed linear electrical resistances with respect to light irradiation. It was noted that these materials deteriorated the optical sensitivity of the microelectrode.

Figure 7 shows the relationship between an electrical resistance of the sensor with an NaCl concentration of 10^{-4} mol l^{-1} in the chamber and the thickness of various kinds of upper board materials [light intensity; (a) 0, (b) 1.44×10^4 Lux]. When the electrolyte concentration is constant, the electrical resistance converges to a constant value that depends on the thickness of all upper boards regardless of the light irradiation [Figures 7(a) and (b), dotted line].

These materials show no response for light intensity. However, the electrical resistance of the sensor increased for upper boards of TF, PP and PCV [□ plot]; in these cases, electrical resistance was reduced when light intensity was high [Figure 7(b)]. These materials show responses for light intensity; thus, packaging materials are classified into two groups in accord with their influence

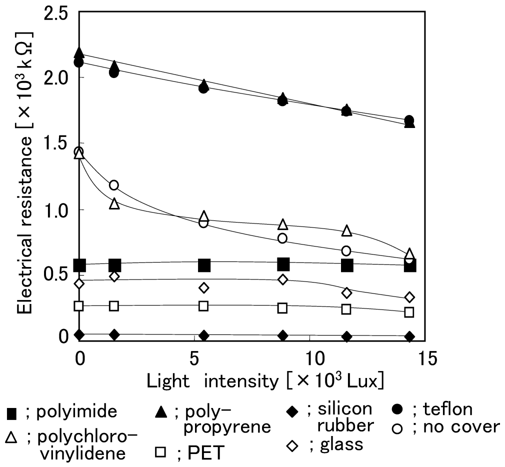

Fig. 6 Relationship between the electrical resistance of microsensors packaged with various materials and the intensity of light irradiation onto the sensors

NaCl concentration, 10^{-4} mol l^{-1}.

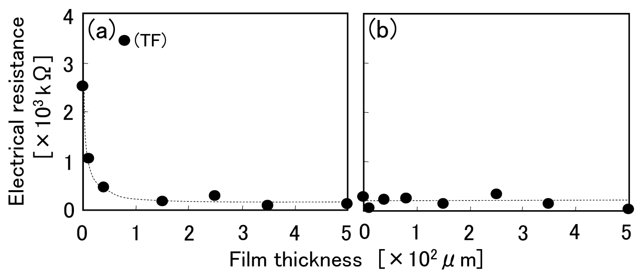

Fig. 5 Relationship between the electrical resistance of the microsensor and the film thickness of the upper board

NaCl concentration; (a) 10^{-4} mol l^{-1}, (b) 1 mol l^{-1}, light intensity; 500 Lux.

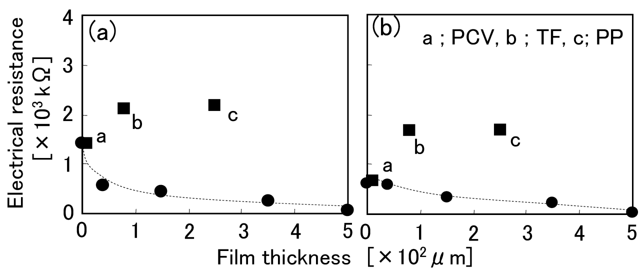

Fig. 7 Relationship between the electrical resistance of the microsensor and the film thickness of the upper board

Light intensity; (a) 0 Lux, (b) 1.44×10^4 Lux, NaCl concentration; 10^{-4} mol l^{-1}.

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on sensor response.

In contrast, when the electrolyte concentration was lower [Figure 2; $10^{-4}\,\mathrm{mol\,l^{-1}}$, Figure 3; $10^{-3}\,\mathrm{mol\,l^{-1}}$, Figure 4; $10^{-1}\,\mathrm{mol\,l^{-1}}$], the decrease in electrical resistance was remarkable. In such cases, the upper board material had a great effect; suggesting that the difference in the adsorption ability of the ion was related to the kind of upper board material. For example, when TF was used as the upper board material [Figure 2], the interaction with the surface was low, because fluorine atoms have a lower reactivity than other elements.

These results demonstrate that photosensitivity and ion sensitivity, as well as the formation of the ion layer, are influenced by choice of the packaging material.

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