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Sensitivity enhancement of mixed-potential gas sensor by a series-connected sensor array

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Abstract:
Mixed-potential gas sensors with an anhydrous proton conductor consisting of zinc metaphosphate glass powder and benzimidazole were fabricated. Reference and sensing electrodes of gold and platinum, respectively, were prepared on an alumina substrate, and a proton conductor was deposited between these electrodes to prepare a gas sensor. The sensor responses to 1-25 ppm H_2 gas in air were investigated. Sensor array with 7 sensors connected in series showed a response to the H_2 gas, even at 1 ppm. The sensitivities of the single sensor and sensor array were −21 and −130 mV/decade, respectively. By connecting 7 sensors in series, the sensor response and sensitivity were improved.

Key words: mixed-potential, gas sensor, hydrogen, phosphate glass, sensor array

Introduction

Mixed-potential gas sensors using yttria-stabilized zirconia (YSZ) electrolyte have been investigated for H_2 detection [1]. These gas sensors must be operated at temperatures above 500 °C to provide both satisfactory response rates against target gases and good gas selectivity. These gas sensors require a heater to maintain the operating temperature above 500 °C, which results in high power consumption. Mixed-potential gas sensors that use electrolytes with high proton conductivity, such as Nafion, zirconium phosphate, and antimonic acid, have been reported to function at reduced operating temperatures [2]. However, these electrolytes must be impregnated with water to achieve high proton conductivity, which necessitates the inclusion of a complicated humidity control system to allow these sensors to operate at room temperature and under high humidity. Moreover, these gas sensors show low selectivity for H_2 against CO. One approach to overcome this challenge is to use anhydrous proton conductors at an intermediate temperature between 100 to 300 °C. However, to the best of our knowledge, no mixed-potential gas sensors using a high-proton-conductivity electrolyte at intermediate temperatures have been reported.

We reported a mixed-potential gas sensor using an anhydrous electrolyte consisting of zinc metaphosphate glass and benzimidazole [3]. The sensor showed good sensor response to H_2 in the concentration range 250-25,000 ppm in both humid and dry air at 120 °C. In this work, it was demonstrated that the sensor response could be enhanced by connecting the sensors in series.

Experimental

A glass with a nominal molar ratio of ZnO-P_2O_5 was prepared by melting a batch mixture of the commercially available reagent-grade chemicals ZnO and H_3PO_4 in a platinum crucible at 1100 °C in air for 30 min. The melt was poured onto an iron plate and quenched by iron pressing to make glass flakes. The glass flakes were milled using an alumina mortar and pestle to below 100 μm in diameter. The glass powder was mixed with benzimidazole, and the mixture was heated at 170 °C for 12 h to make an electrolyte. The weight ratio of the glass powder and benzimidazole was 1:3.

The electrolyte was deposited on an alumina substrate (5 x 15 mm) with platinum (Pt) and gold (Au) electrodes, which have a 1 mm electrode gap and a 1 mm electrode width. The electrolyte-deposited substrate was heated from a back face by a hotplate, and then an electrolyte membrane was formed on the

substrate. We obtained the single sensor and sensor array with 7 sensors connected in series, as shown in Fig. 1.

The sensor element was placed in a test chamber heated to 120°C in an electrical tube furnace. Synthetic dry air (80 vol% N_2, 20 vol% O_2) was introduced into the chamber for 10 min, and a gas mixture of H_2 in dry air was then injected for 10 min. Subsequently, the gas mixture flow was halted and replaced injected at a flow rate of 200 mL/min. The H_2 gas concentration was controlled to the values of 1, 10, 17.5, 25 ppm in dry air.

The potential difference (electromotive force, EMF) between Pt and Au electrodes of the sensor element was recorded using a Potentio/Galvano stat (VersaSTAT4; Princeton Applied Research). During the potential difference measurements, the Pt electrode was always connected to the positive terminal of the Potenio/Galvano stat. The EMFs of the sensor in the air and gas mixtures are denoted as V_a and V_g, respectively. The sensor response (ΔEMF) is defined as $\Delta EMF = V_g - V_a$.

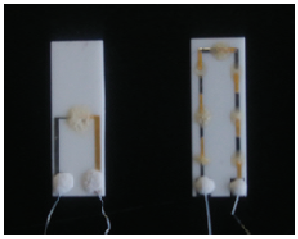

Fig. 1. Optical image of single sensor (left) and sensor array (right).

Results and discussion

Fig. 2 shows the response of the single sensor to different concentrations of H_2 from 1 ppm to 25 ppm in air at 120°C. No sensor response to 1 ppm H_2 was shown. The ΔEMF decreased and reached to saturation values during flowing of H_2 gases for less than 5 min. The ΔEMF increased with the H_2 gas concentration.

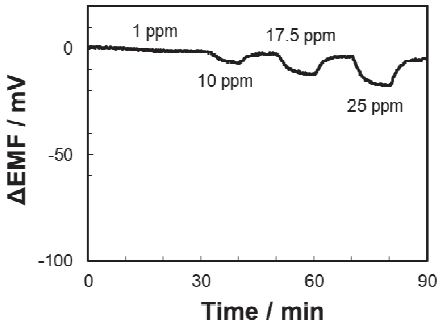

Fig. 2. Response of the single sensor to different concentrations of H_2 from 1 to 25 ppm in dry air at 120 °C.

Fig. 3 shows the response of the sensor array to different concentrations of H_2 from 1 ppm to 25 ppm in air at 120°C. The sensor array showed a distinct response to the H_2 gas, even at 1 ppm. Fig. 4 shows relationship between sensor response and the log of the H_2 concentration for single sensor and sensor array. ΔEMFs were found to be linear to the log of H_2 concentration. The sensitivities of the single sensor and sensor array were −21 and −130 mV/decade, respectively. The sensitivity of sensor array was roughly seven times as compared to that of single sensor.

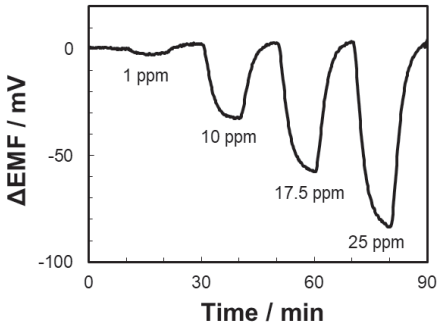

Fig. 3. Response of the sensor array to different concentrations of H_2 from 1 to 25 ppm in dry air at 120 °C.

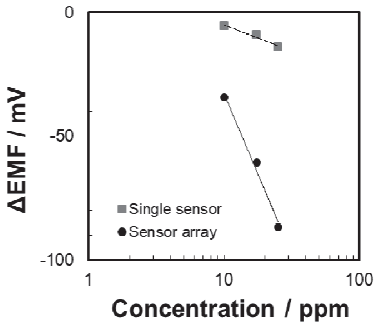

Fig. 4. Relationship between ΔEMF of single sensor and sensor array and H_2 gas concentration.

Acknowledgement

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