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Development of an antenna coil type sensor device for hydrogen leakage detection

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ABSTRACT

An antenna coil type hydrogen gas sensor device was fabricated and evaluated for applying to radio frequency identification (RFID) tag which can easily realize highly reliable and multipoint hydrogen leakage detection system. An antenna coil composed of platinum thin film was formed by photolithography onto a quartz-glass substrate. Then, a platinum-supported tungsten trioxide thin film was deposited on that coil by spin coating method. A frequency dependence of electrical properties (*S*-parameters) of the sensor device in air and in 4 vol.% hydrogen balanced with nitrogen gas was investigated in the range from 300 kHz to 3 GHz. The considerably change in frequency dependence of *S11* with the exposure to hydrogen gas was observed. The result indicated high sensitivity of this sensor device and the possibility of wireless transmission of the information about hydrogen leakage. The 90% response time was about 3 s at 188 MHz. The sensitivity to hydrogen in air was lower than that in nitrogen atmosphere. It could result from the competitive oxidation reaction of tungsten bronze by oxygen. However, the sensor device was able to detect hydrogen gas in concentration range below lower explosion limit. In addition, the change in *S21* between the sensor device and a conventional antenna coil was also demonstrated.

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1. Introduction

From a standpoint of environmental conservation, hydrogen energy system is highly expected because this system does not emit CO_2 gas substantially. Massive infrastructures composed of hydrogen energy system, such as electrolysis plant, liquid hydrogen tanker, large-scale storage tank, long pipeline and fuel cell power plant, are going to develop in the future. However, hydrogen is known as explosive material that has a large combustion range and small ignition energy. Therefore, reliable and low-cost hydrogen gas sensors that can detect leakage of hydrogen gas over a wide area are necessary for handling vast amount of hydrogen gas safely [1].

Until now, two types of hydrogen sensitive sensors, metal-oxide-semiconductor [2] and electrochemical [3] one, were developed and used practically. A high temperature operated oxide-semiconductor gas sensor has high sensitivity, reliability and maintenance-free nature. However, this sensor consumes large electric power for device heating. On the other hands, an electrochemical gas sensor consumes very little power, though electrolyte liquid within the cell has to be periodically replaced for the reliable sensing performance. In addition, these are zero-dimensional type sensors that measure hydrogen gas leakage only at a single point. Therefore, many sensor devices are needed for the leakage monitoring over wide area. Further improvement regarding to operation temperature, maintenance performance and cost are also required.

The wireless sensor networks have been considered as one of the method that are effective in monitoring environment over wide area by distributing small sensors at many measuring points. Wireless sensing is usually classified in active and passive sensing system. Active sensing system needs batteries with limited lifetime. Passive wireless sensors not only have the advantage of being wireless but can also work without batteries. For example, surface acoustic wave (SAW) is one of the passive devices that have been widely investigated and are commonly used for the detection of hazardous compounds in atmospheric environments with high resolution [4]. Fu [5] developed an integrated passive impedance-loaded SAW H_2S sensor. Josse [6] investigated guided shear horizontal surface acoustic wave (guided SH-SAW) devices on $LiTaO_3$ substrates for high-sensitivity chemical and biochemical sensors in liquids. However, these sensor devices require dedicated operating and signal processing electric circuit. These characteristics lead to high cost of device manufacturing. Many other sensor devices also have similar problem.

Radio frequency identification (RFID) devices have simple structure with low cost. In the case of a passive RFID tag, they are only composed of memory microchip and an antenna coil. The antenna coil can be wirelessly transmitted and received information in the

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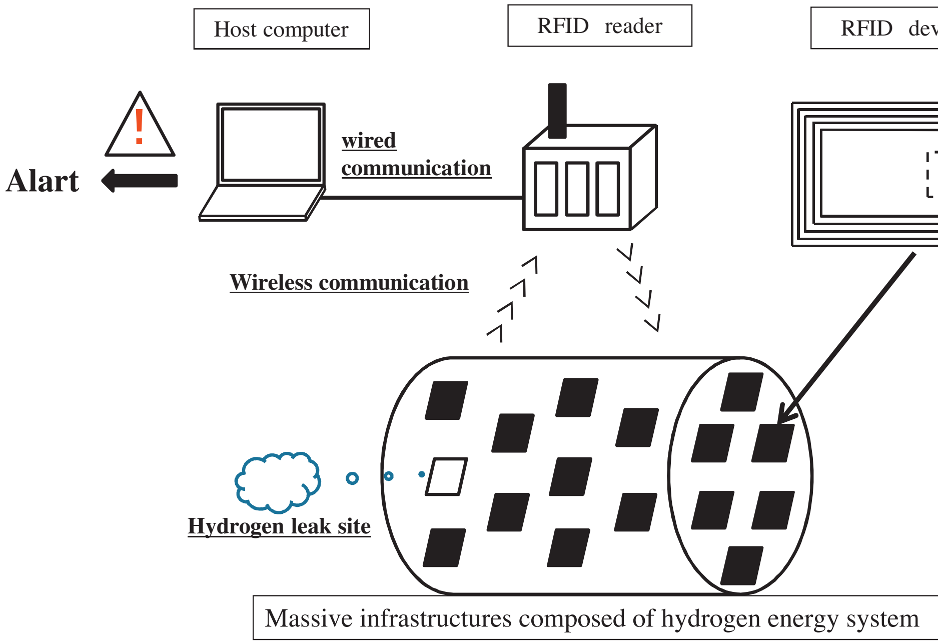

Fig. 1. A schematic diagram of RFID system for hydrogen leakage monitoring.

microchip at specific frequency. The cost of passive RFID tags that operate at 13.56 and 915 MHz has dropped from \$0.5–0.7 in 2003, to \$0.04–0.4 in 2005, and to \$0.02–0.05 in 2006 [7,8]. For having these merits, the RFID system has been used in various fields recently. In the supply chain and food industry, the shipping information has been managed with RFID tags [9]. RFID also has been used for chemical sensing. Potyrailo [10] demonstrated selective vapor sensors that involve the combination of organic electronic sensing materials and passive 13.56 MHz RFID device with multivariable signal transduction. They also applied these sensors to biopharmaceutical industry that required monitoring method of critical manufacturing parameters [11].

In this study, we tried to develop an antenna coil device which has hydrogen sensing function. In our previous studies, we had developed hydrogen sensitive film composed of platinum and tungsten trioxide (WO_3) by using sol–gel method. It was found that the electrical properties of this film drastically changed in the presence of hydrogen gas. In addition, the film showed excellent selectivity and long-term stability [12]. Therefore, we coated sensing film onto antenna coil which has similar structure used in conventional RFID devices. It is supposed that simple and low-cost wireless hydrogen sensor device could be achieved without major design change of coil pattern. Therefore, this sensor will be easily applicable to RFID tag system. As shown in Fig. 1, conventional RFID tags which are coated with the sensing film, are allocated many points to monitor hydrogen leak sites. Then, RFID reader collects these data and sends the information to host computer that issue an alert.

The hydrogen leakage detection capability of antenna coil device was evaluated by measuring the electrical properties at high frequency range for the wireless communication application.

2. Materials and methods

2.1. Sensor fabrication

Fig. 2(a) and (b) shows a prototype antenna coil hydrogen sensing device coated with Pt/WO_3 film. The platinum antenna coil with a size of 45 mm × 45 mm × 400 nm (10 coil turns) was fabricated on the quartz glass plate (800 mm × 800 mm, $t = 1$ mm) using the photolithography. The thickness of antenna coil, which was determined with surface roughness mater, was about 400 nm. The 50 nm titanium layer was deposited on the glass substrate as intermediate layer for enhancing adhesion strength of the platinum coil layer. The impedance characteristics of antenna coil portion were

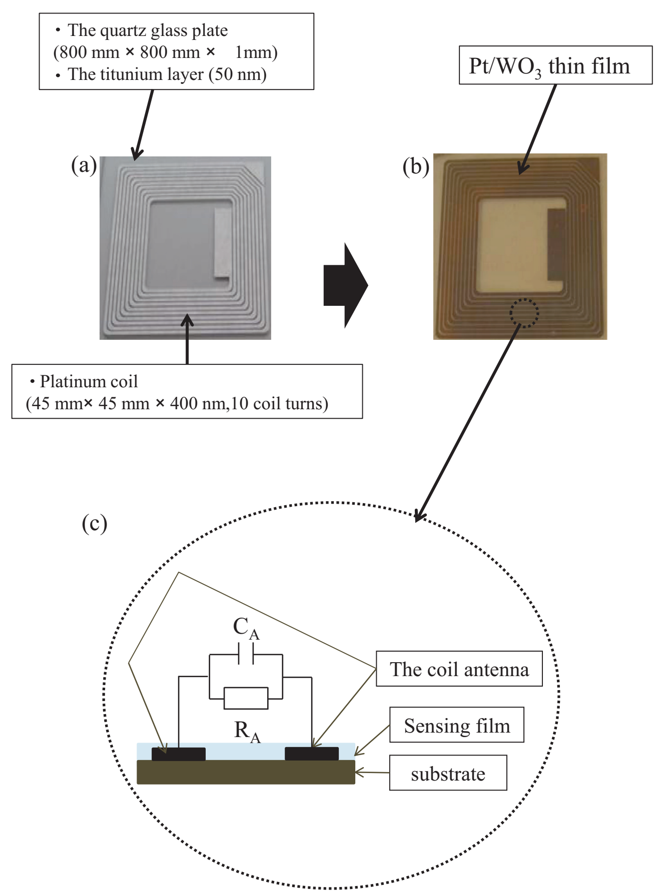

Fig. 2. Structure of an antenna coil type sensor device (a), the sensor device coated with Pt/WO_3 thin film (b) and schematic cross sectional view with the equivalent circuit model of gap between coil patterns (c).

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