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Ultrasonic Hydrogen Sensor: Effects of Environmental Changes

超音波を用いた水素センサー: 環境変化による影響

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1. Introduction

Recently, using hydrogen as an alternative energy to fossil fuel is very important for clean-energy society that does not emit CO_2, NOx, hydrocarbon materials and etc. Hydrogen fuel research has been done for many years and it is known that hydrogen is dangerous gas because it explodes when included more than 4% in the atmosphere. Therefore, it is necessary to develop a faster detection speed sensor to detect leakage of hydrogen below 4%. Variety of hydrogen sensors have been studied for practical application, such as electrochemical sensors, catalytic sensors, solid state sensors, thermoelectric sensors and etc.

Among these sensors, hydrogen sensors using ultrasonic has been reported as a “msec” fast response time sensor [1-3]. This ultrasonic sensor can be affected by environmental changes such as temperature, humidity, and atmospheric pressure. In this study, we have investigated carefully about humidity and temperature changes. We have tried to extract the hydrogen concentration by calculation from the sound speed difference between air and hydrogen contained air in various temperature and humidity conditions.

2. Theory and Experiment

Speed of sound v in the gas with molecular weight of M can be expressed as

$$v=\sqrt{\frac{kRT}{M}} \quad (1)$$

where k is the specific heat ratio, R is the gas constant and T is the temperature. For practical use of the ultrasonic hydrogen sensor, we need to know the temperature and relative humidity of the detecting air. Molecular weight of the moist air can be determined as following.

Saturation vapor pressure E can be expressed as

$$E=6.11\times 10^{\left(\frac{7.5t}{t+237.3}\right)} \quad (2)$$

where t is temperature in Celsius. Also, water vapor pressure Ep can be expressed as

$$E_p=E\times\frac{RH}{100} \quad (3)$$

where RH is relative humidity.

Molecular weight of the moist air Mt can be expressed as

$$M_t=\frac{1013-E_p}{1013}\times M_{da}+\frac{E_p}{1013}\times M_{H_2O} \quad (4)$$

The dry air molecular weight M_{da} is 28.96 g·mol^{-1}, that is calculated from the nitrogen, oxygen and argon ratio in air. The molecular weight of vapor M_{H_2O} is 18.01 g·mol^{-1}

The sound velocity formula can be expressed in case of mixing the hydrogen into the air as following.

$$v_H=\sqrt{\frac{kRT}{M_t(1-\rho)+2.016\rho}} \quad (5)$$

v_H is the sound velocity of hydrogen-mixed air. Hydrogen molecular weight is 2.016 g·mol^{-1} and ρ is hydrogen concentration in volume ratio.

When the speed of sound is v, and a distance is x, the traveling time can be expressed as $t=x/v$. As the hydrogen is mixed in the air, the speed of sound v_H becomes faster. It is expressed using Δt, the arrival time difference between the moist air and the hydrogen-mixed moist air. As a result, v_H can be expressed as eq. (6).

$$v_H=v+\Delta v=\frac{x}{t-\Delta t}=\frac{tv}{t-\Delta t} \quad (6)$$

By using eq. (5) and eq. (6), the hydrogen concentration ρ in the air can be calculated. The following equation shows the relation of the hydrogen concentration ρ and Δt. [1-3]

$$\rho=\frac{M_t}{M_t-2.016}\{1-(\frac{t-\Delta t}{t})^2\} \quad (7)$$

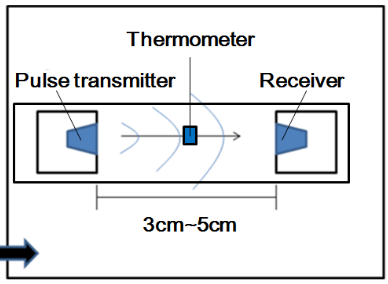

Fig.1 Diagrammatic illustration of an ultrasonic hydrogen sensor device.

We prepare the sound speed measuring system as show in **Fig. 1**. Two standard ultrasonic probes were placed in a 5L air tight box for transmitting and receiving the pulse. A thermometer was placed between the two probes to measure the temperature between probes. The sound travelling time was measured when the temperature was varied from -10 ºC to 50 ºC. We compared the experimental and theoretical expression of Δt between dry air and moist air in order to confirm the theory of humidity indicated earlier. For all the calculation Δt was set as zero at 20 ºC with no hydrogen. When we measured the temperature and humidity, this air tight box was placed in a thermostatic bath until the end of the experiment, so that absolute humidity did not change. The air in the box was introduced at room temperature before placing into the thermostatic bath.

3. Result and discussion

Figure 2 is theoretical curves of Δt for the dry air and for the moist air (*Ep*=18.7hPa) for different temperature. The Δt is the arrival time difference between 20ºC (dry air) and each temperature of the dry air or the moist air (*Ep*=18.7hPa). A Solid line is a theoretical curve of the moist air (*Ep*=18.7hPa), and a dashed line is a theoretical curve of the dry air. According to eq. (2)-(5), the molecular weight decreases when the humidity is considered, therefore; the speed of sound becomes faster and thus Δt slightly increases.

Figure 3 is theoretical and experimental values of Δt for different temperature in the moist air (*Ep*=10.4hPa). A solid line is a theoretical curve, and dots are the experimental values. Good fitting was obtained between theoretical and experiment values. Thus, it appears to be possible to calibrate for the hydrogen concentration with the temperature and humidity variation.

Figure 4 shows hydrogen concentration calculated from the measured Δt according to eq. (7). About 3.5 vol% of hydrogen concentration was injected in the air tight box which contained the moist air (*Ep*=24.8hPa).

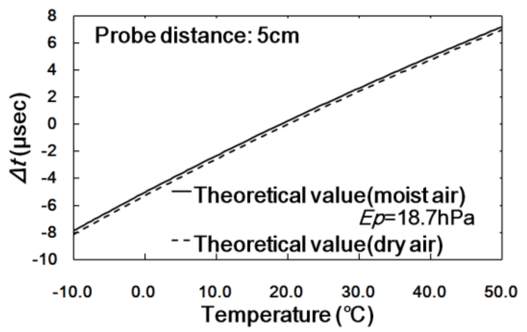

Fig.2 Theoretical values of Δt for different temperature. A Solid line is for the moist air (*Ep*=18.7hPa), and a dashed line is for the dry air.

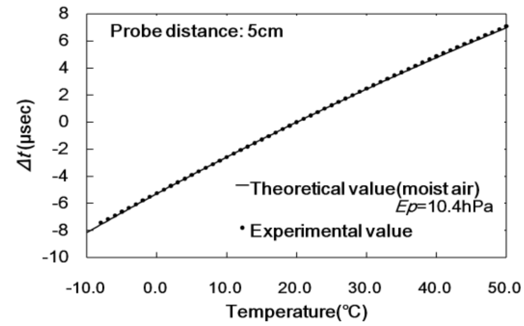

Fig.3 Theoretical and experimental values of Δt for different temperature in the moist air (*Ep*=10.4hPa). A solid line is a theoretical curve, and dots are the experimental values.

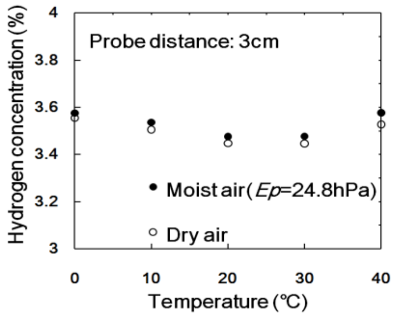

Fig.4 Measured hydrogen concentration derived from eq. (7). Δt was actually measured values.

It appears that differences in hydrogen concentration due to the temperature change are less than ±0.1%. For this experiment hydrogen was injected in the box when it reached at each temperature, therefore; there are slight human errors on injecting hydrogen. Also, the difference in the calculation between the dry air and the moist air (*Ep*=24.8hPa) is less than 0.05%. Therefore, it can be concluded that humidity correction can be neglected for hydrogen concentration when the measurement accuracy is allowed to be about 0.1%.

4. Conclusion

In this study we have shown that using ultrasonic, hydrogen concentration can be measured in temperatures range of -10ºC to 40 ºC in the moist air. The difference in the calculated hydrogen concentration between the dry air and the moist air (*Ep*=24.8hPa) can be less than 0.05% and may be neglected for practical use.

References

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