

# Measurement of a wide range of hydrogen concentration with rapid response using dual pressure gauges

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#### ABSTRACT

We investigated the dependence of hydrogen sensing on the hydrogen density at 0.1–100 vol% in air atmosphere by commercially available diaphragm and quartz friction pressure gauges (D- and Q-gauges). It was demonstrated that the Q-gauge pressure reading depends on the hydrogen concentration for concentrations up to 100 vol% and can be used to measure the hydrogen concentration in air. The response times using the dual pressure gauges were shown to be below one second for hydrogen concentrations of 0.1–100 vol% in air atmosphere, indicating that our hydrogen-sensing method has a rapid response in the above range of hydrogen concentrations. In conclusion, hydrogen sensing with dual pressure gauges can be used to measure a wide range of hydrogen concentrations with a fast response; thus, this method is particularly practical for ensuring a safe hydrogen-based economy.

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### 1. Introduction

Hydrogen sensing requires technology that enables the following points to be realized: i) a small lowest detection limit for hydrogen concentration, ii) a wide measurement range for the hydrogen concentration, iii) reproducibility of the measurement, iv) a fast response time for detecting hydrogen leakage, v) applicability for various environments for hydrogen-based economy including outdoors, vi) a wide range of operating temperatures and humidity, vii) interference resistant against various gas environments, viii) a long life-time of the measurement device, and ix) safe detection

without the consumption of hydrogen and energy such as heat, electricity, or photons. Some of the specifications above for hydrogen sensing have also been announced by the U. S. Department of Energy (DOE) [1]. Until now, there are several types of hydrogen sensors to be studied such as semiconductor [2–7], thermoelectric [8], optical [9], thermally conductive [10], and other noble sensors [11–14]. Oxide based semiconducting sensors work through the change in resistivity or surface conductivity and have relatively lower detection limit [2]. Thermoelectric (TE) sensors consist of the catalysts which enhance the oxidation of hydrogen and the thermoelectric materials which convert the heat generated by

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the reaction with hydrogen to the electric output [8]. Advantages of TE sensors are high reliability, environmental-free, and their outputs are mostly linear to the hydrogen concentration. Optical sensors monitor the change in reflectance or transmittance by the hydrogen absorption with fast response [9]. Thermal conductive sensors measure the gas composition change by the membrane temperature change and particularly suitable to the higher hydrogen concentrations [10].

However, no hydrogen sensors have been reported that meet all the requirements listed in i)-ix) above in a single device. For example, although the lowest detection limit for hydrogen concentration for a pseudomorphic high electron mobility transistor is 4.3 ppm in air at room temperature, its response time is on the order of 10 s [2]. One of the fastestresponse hydrogen sensors are the palladium mesowire arrays (PMAs), which detect low hydrogen concentrations within 20 ms but have a longer response time for higher hydrogen concentrations [15]. Also, the sensitivity of PMAs to concentrations above 10 vol% is relatively poor, and they are difficult to use for measurements of higher hydrogen concentrations. On the other hand, 3C-SiC sensors show a response to hydrogen concentrations in the range of 0.33-100 vol%; however, their response times are on the order of minutes [3]. For greater safety and lower cost, it is more practical to detect high H<sub>2</sub> concentrations using one sensor. Simultaneous quick H<sub>2</sub> concentration measurements also should be more convenient to ensure the detection of H<sub>2</sub> leakage. It is reported that few sensors have a response time below one second. In addition, most of the sensors studied need heating to enhance the sensitivity.

We have previously reported that hydrogen leakage and concentration changes can be detected using commercially available pressure gauges of a quartz friction pressure gauge (Q-gauge) and a diaphragm gauge (D-gauge) [16]. This measurement is based on the principle that the pressure measured using a Q-gauge (hereafter Q-gauge pressure) depends on not only the absolute pressure but also the molecular weight and the viscosity of the gas [17,18]. Since there are large differences between the molecular weight and viscosity of  $H_2$  (2.02, 8.35  $\mu$  Pas) and air (28.97, 17.08  $\mu$  Pas), one can detect the hydrogen leakage by measuring the viscosity and the absolute pressure [19]. Our sensing method can detect 0.05–10 vol%  $H_2$  in air with a response time of 600 ms at atmospheric pressure [16]. The present Q-gauge measurement detects the hydrogen through the pressure measurement, therefore; its response time should be comparable to the response times of the pressure gauges, which are on the order of several hundred milliseconds at the maximum, independent of the H<sub>2</sub> concentration in air. This rapid response of the gauges results in the rapid detection of  $H_2$  leakage and the measurement of the  $H_2$  concentration. Another merit of this method is safe sensing because hydrogen leakage is detected by measuring the pressure of the gas without the use of any energy or elevating temperature. A small quartz sensor with size less than 1 cm makes it possible to downsize the sensing equipment. Thereby, the sensing device can be applied to the various installations necessary for a hydrogen economy such as those in the production, storage, and distribution of hydrogen, and in fuel cell vehicles [17].

In this study, the  $H_2$  concentration range measurable using dual pressure gauges was investigated. Moreover, the response times for  $H_2$  detection were also measured at various  $H_2$  concentrations. The results given in this study show that our hydrogen-sensing method is practically useful for ensuring safety in a hydrogen economy.

### 2. Experimental

### 2.1. Outline of setup

Experimental setup used in this study is the same as that used in our previous study [16,17]. Briefly, the setup consists of a test chamber and gas supply lines. The test chamber is a 90 cm<sup>3</sup> stainless-steel tube with a Q-gauge (VP Co. model GC-210) and a D-gauge (Baratron model 222BA) and is connected to manual gas supply valves to introduce H<sub>2</sub> and air. Electric valves for gas switching, which can be used up to 10 MPa, were employed to measure the response time to H<sub>2</sub> leakage. Compressed air from the atmosphere was used for the source for the air gas without any further purification. On the other hand, the source of  $H_2$  gas was a cylinder (purity: >99.99999%). Both gases were introduced from each source into the test chamber through the mass flow controllers (MFCs) which control each gas flow. In this study, the Q-gauge was calibrated by air, which means that the Q-gauge pressure reading gives the absolute pressure when pure air is measured. All measurements in this study were carried out at room temperature under air-conditioned atmosphere kept to 25 °C. Relative humidity (RH) in the test chamber was below 5RH% during the experiments all the time. In addition, the readout from the Q-gauge used in this study was automatically temperature calibrated. Thereby, the influences of temperature and humidity are negligibly small except the investigation on the temperature and humidity dependences.

# 2.2. Measurement to obtain working curve using static gases

To obtain the  $H_2$  concentration dependence of the Q-gauge reading, the Q- and D-gauges were used to measure the pressure in the chamber while changing the partial pressure of  $H_2$  in air at 100 kPa. Gas mixtures with known concentrations of  $H_2$  in air were prepared by introducing  $H_2$  and air through manual inlet valves into the test chamber while closing the outlet valve fixed on the vacuum pump [16,20]. The concentrations of air and  $H_2$  were adjusted while measuring the absolute partial pressure of each gas by the D-gauge. Details of the production of these gas mixtures will be presented in Section 3.1. After that, the pressure of the chamber was measured using the dual pressure gauges for each static gas mixture.

#### 2.3. Measurement for response times using gas flows

The response time to  $H_2$  with various concentrations in air and the recovery time from  $H_2$ -contaminated air to pure air were investigated using gas flows. For these time response measurements,  $H_2$  leakage into air was simulated by swapping the pure air with a mixture of  $H_2$  and air. Because the order of the response time is supposed to be less than one second, the exchange between pure air and the gas mixture must be done in less than several tenths of a millisecond. The gas switching between pure air and the mixture of  $H_2$  and air was done using "switching valves (SWV)", which consists of four electric operating valves [16]. Premixing between  $H_2$  and air was also carried out by the connection of the controlled gas flow supplies of  $H_2$  and air. This also reduces the time required for the measurement and allows us to derive the intrinsic response time of our sensing method because the mixing process is also one of the rate-determining steps in the response time measurement.

Rapid gas switching using SWV was performed at the opening and closing times of each diaphragm electric valve of 5 and 3.5 ms, respectively, which are sufficiently fast for the present measurement of response times. Moreover, these valves were connected as closely as possible to each other to reduce the time required for gas switching. The SWV was connected to the two inlet and two outlet tubes and was connected as closely as possible to the Q-gauge. One of the outlets is the test chamber and the other is for a bypass pump. Before, during, and after the gas exchange between air and air-H<sub>2</sub>, the total pressure always needed to be kept constant at 100 kPa. Because the Q-gauge pressure depends on the total pressure, a sudden marked change in total pressure may affect the response time. To make it easier to adjust the total pressure in the test chamber during the gas switching, both air and air-H<sub>2</sub> were flowed. To ensure the continual flow of gases in the tubes, another outlet of the bypass pump for the gases that do not flow into the test chamber was installed. The total pressure at the test chamber was controlled to 100 kPa by adjusting the manual regulating valves.

As a result, gas switching between pure air and the  $H_2$ -air mixture was achieved within 50 ms while maintaining the total pressure at 100 kPa in the test chamber before and after the gas switching. The flow rates of air before switching, and  $H_2$  and air after switching were varied in the ranges of 100–140, 1.4–4500, and 0–240 standard cubic centimeters per minute (sccm) using the MFCs, respectively, to measure the response times for various  $H_2$  concentrations in air. Temporal measurements of the Q- and D-gauges were carried out with a time resolution of 50 ms. Influences of the change in temperature and humidity can be neglected during the response time measurement because the necessary time for measuring one set of response and recovery time on the hydrogen leakage was about 30 s.

# 3. Results and discussion

### 3.1. H<sub>2</sub> leakage detection using D- and Q-gauges

The temporal changes of both Q- and D-gauge pressure readings after closing the outlet valve and the introduction of both gases of 99.8 kPa air and 0.1 kPa  $H_2$  are shown in Fig. 1(a). Each gas was introduced while measuring the absolute pressure by the D-gauge. In this case, after the evacuation of the chamber, the outlet valve on the vacuum pump was closed. At that time, the D-gauge pressure was calibrated to zero. Then



Fig. 1 – Temporal changes of Q- and D-gauge pressure readings [(a),  $\circ$ ,  $\bullet$ ] and normalized pressure readings from Q- and D-gauges [(b),  $\Box$ ,  $\blacksquare$ ] when 0.1 kPa H<sub>2</sub> was introduced into 99.8 kPa pure air with the outlet of the chamber closed.

air was introduced until the D-gauge pressure reached 99.8 kPa, meaning that the partial pressure of air was 99.8 kPa in the chamber. Next, in the same way,  $H_2$  was introduced until the D-gauge pressure indicated 99.9 kPa. As a result, the difference between the D-gauge pressures before and after the introduction of  $H_2$  is 0.1 kPa, corresponding to the partial pressure of  $H_2$  in this gas mixture.

During the mixing of both gases after their introduction, the D-gauge reading indicated an almost constant pressure of about 100 kPa; on the other hand, the Q-gauge pressure changed over several minutes. Since the absolute pressure is almost constant and the chamber is closed, the change in the Q-gauge pressure is not due to the change in total pressure but to the change in the molecular weight and viscosity of the gases in the chamber. The change in the Q-gauge reading means that it takes several minutes for the mixture of air and H<sub>2</sub> to reach a uniform concentration in the chamber.

Finally, the Q-gauge indicated a pressure reading of lower than 100 kPa at 10 minutes after the mixing. These changes were qualitatively identical for  $H_2$  concentrations of 0.1–100 vol%, indicating that there are no reactions at room temperature between air and  $H_2$  in the test chamber. The decrease in the Q-gauge pressure reading, which indicates the decrease in the average molecular weight and viscosity of the gas mixture to be measured, agrees with the changes in the molecular weight and viscosity that occur after the introduction of  $H_2$  into air. Thereby, it was shown that hydrogen sensing is possible using dual pressure gauges consisting of Q- and D-gauges.

To improve the stability of the detection of hydrogen leakage over a long time range, fluctuations of the atmospheric pressure need to be cancelled. To achieve this, the ratio of the Q-gauge pressure to the total pressure measured by the D-gauge was employed. This pressure-normalized Qgauge pressure and the normalized total pressure are plotted in Fig. 1(b). The normalized total pressure was obtained by normalization using the initial (time = 0 s) total pressure measured by the D-gauge. Independent of the total pressure, the normalized Q-gauge pressure in Fig. 1(b) showed more rapid saturation than the Q-gauge pressure in Fig. 1(a). This shows that the influence of unstable atmospheric pressure can be excluded using the ratio of the Q-gauge pressure to the D-gauge pressure. After this, we call this ratio the "Q-gauge ratio". The fluctuation of the Q-gauge reading was about 0.1 kPa, which determines the minimum detectable H<sub>2</sub> concentration by our sensing method, which will be discussed in detail in next subsection.

# 3.2. H<sub>2</sub> concentration measurement and minimum detectable concentration

Fig. 2 shows the H<sub>2</sub> concentration dependence of the Q-gauge ratio at 100 kPa. Each H<sub>2</sub>-air mixture with a different hydrogen concentration was prepared by measuring the absolute pressure of each gas with the D-gauge using the method discussed in Section 3.1. The Q-gauge ratio depends on the viscosity and molecular weight of the gas mixtures because the influence of the total pressure was excluded by using the Q-gauge ratio [18]. As seen in Fig. 2, it was found that the Q-gauge ratio decreases with increasing H2 concentration in air for concentrations of 0.1-100%, indicating that the Q-gauge ratio depends on the H<sub>2</sub> concentration for concentrations up to 100 vol%. The dependence of the Q-gauge ratio on the H<sub>2</sub> concentration qualitatively agrees with the changes in the molecular weight and viscosity of the gas mixture since both the averaged molecular weight and the viscosity of the gas mixture decrease with H<sub>2</sub> concentration. This can be



Fig. 2 – Hydrogen concentration dependence of Q-gauge ratio ( $\bullet$ ). The estimated relative viscosity of the gas mixture of H<sub>2</sub> and air at 100 kPa is indicated by a line calculated by Wilke's formula.

explained by the change in friction applied to the quartz sensor involved in the Q-gauge [21]. Using the working curve in Fig. 2, one can measure  $H_2$  concentrations of up to 100 vol% in air using the dual pressure gauges at a constant pressure of 100 kPa.

The accuracy of the present H<sub>2</sub> concentration measurement can be estimated from the fluctuation of the Q-gauge ratio in Fig. 1(b) and the working curve in Fig. 2. The Q-gauge ratios at 0 and 100 vol% are 1.0 and 0.024, respectively. Therefore, the decrease in the Q-gauge ratio for each 1 vol% H<sub>2</sub> concentration is (1.0-0.024)/100 = 0.0098. From Fig. 1(b), the fluctuation of the present measurement was 0.001; therefore, the accuracy of the present measurement is estimated to be 0.1 vol%. This indicates that the lowest detection limit for hydrogen is 0.1 vol% and is sufficiently low for the detection of a small hydrogen leak. These results indicate that this method of hydrogen sensing can measure a wide range of hydrogen concentrations of 0.1-100 vol% at 100 kPa. Using this method, one can determine the concentration of  $H_2$  leakage when a leak is detected. This simultaneous measurement of the concentration of H<sub>2</sub> is advantageous in terms of safety.

# 3.3. Viscosity of air–H<sub>2</sub> gas mixture estimated from Q-gauge measurement

To understand the  $H_2$  concentration dependence of the Qgauge ratio in Fig. 2 in detail, we discuss the viscosity of the mixture of air and  $H_2$  in this section. The Q-gauge ratio depends on the molecular weight and viscosity of the gas mixtures, and is not always linear against the  $H_2$  concentration because the partial pressure dependence of the viscosity of a binary gas mixture is not necessarily linear and depends on the combination of gases [22]. In the viscous flow region at 100 kPa, the Q-gauge ratio can be expressed as follows:

$$\mathbb{R} \propto \varDelta Z^2 \propto \eta \rho \propto \eta M,$$
 (1)

where R,  $\Delta Z$ ,  $\eta$ ,  $\rho$ , and M are the Q-gauge ratio, the impedance change of the quartz sensor in the Q-gauge, viscosity, density, and molecular weight of each gas in the gas mixture, respectively [21,23].

In the case of air and  $H_2$ , since there are no data reported to our knowledge, we estimated the  $H_2$  concentration dependence of the viscosity of the mixture of air and  $H_2$  using Wilke's form as follows [22].

$$\eta_m = \sum_{i=1}^n \frac{\eta_i}{1 + \frac{1}{x} \sum_{\substack{j=n \\ J \neq i}}^{j=n} x_j \phi_{ij}}.$$
(2)

Here,

$$\phi_{ij} = \frac{\left[1 + \left(\eta_i/\eta_j\right)^{1/2} \left(M_j/M_i\right)^{1/4}\right]^2}{\left(4/\sqrt{2}\right) \left[1 + \left(M_i/M_j\right)\right]^{1/2}} \cdots.$$

For binary gas mixtures, i = 1 and j = 2. Using Eq. (1), one can derive the viscosity of a binary gas mixture from  $\eta$  and M for pure gases that comprise the mixture. The molecular weight of air was assumed to be 28.8 from the composition with 80 vol% N<sub>2</sub> and 20 vol% O<sub>2</sub>.

By substituting Eq. (1) in Eq. (2), we can compare our experimental results of R with the viscosity of the mixture of

air and  $H_2$  obtained from Wilke's formula by normalization with the viscosities of 100 vol% air and 100 vol%  $H_2$ . The ratios of the calculated viscosity of the mixtures of air and  $H_2$  are shown in Fig. 2 by a line for comparison, and indicate good agreement with the experimental results. This means that R strongly depends on the viscosity of the gas mixture. Therefore, the above results show that the viscosity of a gas mixture can be obtained from the present Q-gauge setup and the viscosity of each gas that constitutes the gas mixture. The safety of measuring the viscosity of a gas mixture is another advantage of the present measurement, particularly for highly reactive gases.

# 3.4. Response times for $H_2$ leakage detection at various $H_2$ concentrations

Next, we investigated the response times of this hydrogensensing method to the introduction of  $H_2$  and to the reduction of  $H_2$ . The response times of this hydrogen-sensing method have already shown to be below one second for 0.5 vol%  $H_2$ [16]. This sensing method is based on pressure measurement; therefore, the response time should be comparable to the response times of the Q- and D-gauges, which are about several hundreds of milliseconds at the latest. To measure such fast response times below one second, gas switching by the SWV between the pure air flow and the  $H_2$ -air flow was used to reduce the gas switching and the mixing times, as explained in Section 2.3 in detail. The time responses of the Q- and D-gauge pressure readings and the Q-gauge ratio are plotted in Fig. 3(a) and (b) together with the normalized D-gauge pressure reading. About 5 s after the introduction of 101 sccm pure air, the gas flow was switched to a mixed gas flow of 10 sccm  $H_2$  and 147 sccm air using the SWV. The time responses to this gas switching are indicated in Fig. 3 by lines.

As presented in Fig. 3(a) and (b), the Q-gauge pressure and Q-gauge ratio quickly decreased within 500 ms of the introduction of hydrogen to reach 90% of the saturated value. The Q-gauge ratio returned to the value before the introduction of the  $H_2$ -air mixture within about 500 ms when the hydrogen mixed air was switched back to pure air. The saturation of the Q-gauge ratio in Fig. 3(b) after the introduction of  $H_2$  seems to be more rapid than that of the Q-gauge pressure in Fig. 3(a). In addition, there was a smaller difference observed between the Q-gauge ratio in Fig. 3(b) before  $H_2$  introduction and after  $H_2$  reduction than that for the Q-gauge pressure in Fig. 3(a). These results again indicate that it is more precise and stable to use the Q-gauge ratio rather than the Q-gauge pressure.

The response times were measured for various  $H_2$  concentrations up to 100 vol% and some of the results are presented in Fig. 4. The  $H_2$  concentration in each gas mixture was controlled by varying the flow rates of  $H_2$  and air using the MFCs, and was measured using the working curve in Fig. 2 as presented in Section 3.2. Both response times were comparable to the results shown in Fig. 3 for all  $H_2$  concentrations, indicating that both response times are below one second even for the detection of 100 vol%  $H_2$ . The irregular changes in the temporal Q-gauge ratio around t = 7 s after the introduction of  $H_2$  are probably due to the unintentionally stored gas in the spaces in the electric valves or to the difference between the inlet and outlet conductance, or both.



Fig. 3 – Temporal changes of Q- and D-gauge pressure readings [(a),  $\circ$ ,  $\bullet$ ] and normalized pressure readings from Q- and D-gauges [(b),  $\Box$ ,  $\blacksquare$ ] when 101 sccm pure air was switched to a mixed gas flow of 10 sccm H<sub>2</sub> and 147 sccm air then switched back to the flow of 101 sccm pure air.



Fig. 4 – Temporal changes in Q-gauge ratio for various  $H_2$  concentrations of 4–100 vol%.

The H<sub>2</sub> concentration dependence of both response times is summarized in Fig. 5, indicating that the response time of this hydrogen-sensing method is 300-800 ms, independent of the hydrogen concentration. Therefore, it is shown that this sensing method can detect hydrogen leakage with H<sub>2</sub> concentrations of 0.1-100 vol% within one second. The scattering of data is perhaps due to differences of the flow rates of the gases introduced into the test chamber. Consequently, the intrinsic response time of this method might be around 300 ms, independent of the H<sub>2</sub> concentration. These response times on the order of several hundreds of milliseconds are comparable to the intrinsic response times of the Q-gauge, thereby, the response times obtained for the present hydrogen-sensing method are reasonable. The present setup can measure the H<sub>2</sub> concentration at the same time as detecting the H<sub>2</sub> leakage with a response time of several hundred milliseconds. This is a great advantage for the present measurement. These quick response times are also advantageous for installations in a hydrogen economy, because of not only the fast detection of H<sub>2</sub> leakage, but also the fast recovery time from the air-H2 mixture to pure air because the reproducible and repeatable measurements are possible with a fast response time even for higher H<sub>2</sub> concentrations.

### 3.5. Discrepancy between relative $H_2$ concentration and relative $H_2$ flow rate in the gas flow

There is large difference between relative  $H_2$  concentration and relative  $H_2$  flow rate in the gas flow. After the introduction of the  $H_2$ -air mixtures in Fig. 3, the  $H_2$  concentration was 0.15 vol%, as read off from the working curve in Fig. 2, however, this does not agree with the relative flow rate ratio of  $H_2$  to air (10 sccm/147 sccm = 6.8%). In a viscous flow, the speed of each gas is uniform and independent of its mass; therefore, the partial concentration of each gas in the mixture should be identical to the corresponding gas flow. The difference between the  $H_2$  concentration in the gas mixture flow and the flow rate ratio is due to the differences between the molecular weight and viscosity of  $H_2$  and air. Since the

1.0

Response time (sec)

molecular weight and viscosity of  $H_2$  are smaller than those of the other gases,  $H_2$  should be evacuated faster from the chamber than air; therefore, the  $H_2$  concentration was lower than the  $H_2$  flow rate ratio [24]. The  $H_2$  concentration was plotted against the relative  $H_2$  flow rate in Fig. 6, which indicates that the relative  $H_2$  concentration is always smaller than the relative  $H_2$  flow rate.

Note that flows of 10 sccm  $H_2$  and 147 sccm air are those at the MFCs and fixed by the MFCs. This indicates that there are differences in the flow rate ratio depending on the position in the gas tube because the relative  $H_2$  concentration is smaller than the relative flow rate of  $H_2$  at the position of the Q-gauge. Therefore, the  $H_2$  concentration measurement is necessary to obtain the actual flow rate of  $H_2$  in air even when the flow is fixed by the MFCs.

#### 3.6. Influences of temperature and humidity

Hydrogen sensors are often used at outdoor facilities such as hydrogen production and storage facilities, and filling stations. Thus, they need to be resistant to changes in the temperature and humidity. Since the resonant frequency of the quartz in the Q-gauge depends on the temperature and the Q-gauge pressure depends on the frequency, the Q-gauge pressure should depend on temperature [25]. The Q-gauge pressure also probably depends on the H<sub>2</sub>O concentration in air, that is, the humidity. If these changes in the Q-gauge pressure by temperature or humidity are larger than that caused by the introductions of H<sub>2</sub>, it may result in an error in the detection of hydrogen leakage.

To investigate the influences of temperature and humidity, their effects on the Q-gauge pressure were investigated [26]. Experimental results showed that the Q-gauge ratio is proportional to the temperature in dry air and inversely proportional to the absolute humidity at various constant temperatures. This means that the present setup can measure the  $H_2O$  concentration, and can thus also be used as a humidity sensor under a  $H_2$ -free environment. The changes in the Q-gauge ratio over 1 °C and 1 kg dry air were 0.00151 and 0.80, respectively.

Using these values, the change in the baseline of the Qgauge ratio upon an increase in temperature from 25.5 to 29.0 °C and an increase in humidity from 18 to 90% over 1 h



Fig. 5 – Response time to the introduction of hydrogen ( $\odot$ ) and the recovery time ( $\bullet$ ) upon reverting to pure air for various H<sub>2</sub> concentrations of 0.1–100 vol%.



Fig. 6 – Hydrogen flow rate ratio ( $\odot$ ) dependence of hydrogen concentration in flows of H<sub>2</sub> and air.

was reduced to 0.5 kPa from 1.5 kPa. Actual changes in temperature and humidity over 1 h should be smaller than the above; therefore, the sensor still can work normally outside upon temperature and humidity normalization.

#### 3.7. Interferences of other gases

The present measurement system is based on the differences between the viscosity and molecular weight of hydrogen and air; thereby, other gases may also affect the Q-gauge pressure. One such gas is  $H_2O$ , as mentioned in the previous subsection. Other gases with a smaller or larger molecular weight or viscosity than the components in air may affect the Q-gauge pressure because it depends on the types of gas at constant absolute pressures [17]. The Q-gauge pressure dependences on the types of gas is reported for the same type Q-gauge as ours used in this study [17]. Judging from the results in Ref. [17], the differences of the Q-gauge pressure between N<sub>2</sub>, Ar, and Kr at around atmospheric pressure are relatively small. Therefore, Ar and Kr do not disturb the present hydrogen sending measurement using the dual pressure gauges in air  $(N_2)$ , because the introduction of Ar or Kr into  $N_2$ do not affect the Q-gauge pressure. In other words, the gas which does not affect the air-calibrated Q-gauge pressure does not disturb the present measurement. More generally, the leakage of the gases with higher molecular weight or viscosity seems not to interfere in the air-calibrated Q-gauge pressure and our hydrogen measurement.

From this point of view, the gases with smaller molecular weight and viscosity compared to those of air may interfere in the present hydrogen measurement in air as well as  $H_2O$ . To study the influence of such gases, helium concentration dependence for the Q-gauge pressure at 100 kPa was experimentally investigated because its molecular weight (4.00) is small and relatively close to that of  $H_2$ . The Q-gauge ratio measured for mixtures of air and helium at 100 kPa is plotted against the helium concentration in Fig. 7. Compared with Fig. 2, the influence of the helium concentration is smaller than that of  $H_2$ , but the presence of helium probably affects the Q-gauge ratio.

Finally, probable interfering gases for our measurement are  $H_2O$  and He. Their introduction possibly results in the same change by  $H_2$ . Influences of  $H_2O$  can be cancelled out by



Fig. 7 – Helium concentration dependence of Q-gauge ratio (○) in air at 100 kPa.

the simultaneous measurement for the humidity as explained in Section 3.6. Other possible disturbance for the Q-gauge measurement maybe the corrosive gases such as fluorinated gases for the quartz in the Q-gauge. Otherwise, the Q-gauge measurement is not influenced by the gases unless they deposit on the surface of the quartz. Such depositions by contamination should be proceeding temporally slowly; therefore, the Q-gauge measurement may detect the fast hydrogen leakage. Even the surface of the quartz is covered; the Q-gauge measurement should be possible if the oscillating frequencies can be precisely measured.

## 4. Conclusions

We have presented a hydrogen-sensing method using commercial Q- and D-gauges with short response times to  $H_2$ leakage over a wide range of  $H_2$  concentrations. Our measurement methods can detect  $H_2$  with its concentration with 0.1–100 vol% below 800 ms with measuring  $H_2$  concentration simultaneously. It was also found that the Q-gauge ratio correlates to the viscosity of the  $H_2$ -air mixtures. The present  $H_2$  concentration measurement showed that there are large discrepancies between the flow rate ratios and the partial pressure ratios of  $H_2$  to air.

Our Q-gauge measurement meets most of the requirements of hydrogen sensing stipulated by the DOE [1]. By improving the data processing and using a quartz sensor with a smaller fluctuation instead of the Q-gauge, measurement that is more sensitive should be possible. Taking into account the safety measurement using dual pressure gauges, the present measurement has many advantages and promises safer hydrogen sensing in a hydrogen economy.

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