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REVIEW

Principle and application of ball surface acoustic wave (SAW) sensor

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Abstract: Detection of hydrogen gas is a crucial task for establishing safety and reliability of fuel cells, a key technology for the environment and our society. However, hydrogen is difficult to detect and various hydrogen sensors have many drawbacks. Here we report a novel hydrogen gas sensor, the ball surface acoustic wave (SAW) sensor, using Pd or PdNi sensitive film. The ball SAW sensor is based on a novel phenomenon, diffraction-free propagation of collimated beam along an equator of sphere. The resultant ultra-multiple roundtrips of SAW makes it possible to achieve highest sensitivity among SAW sensors. Moreover, it enables to use a very thin sensitive film, and consequently the shortest response time (2 s) was realized. In terms of the sensing range, it has the widest range of 10 ppm to 100% among any hydrogen sensors including FET or resistivity sensors. The response time was less than 1 s for 3.0% hydrogen concentration in nitrogen gas, evaluated by using a newly developed digital quadrature detector.

Keywords: Surface acoustic wave, Bearing ball, Gas sensor, Hydrogen, Naturally collimated beam, Multiple roundtrip

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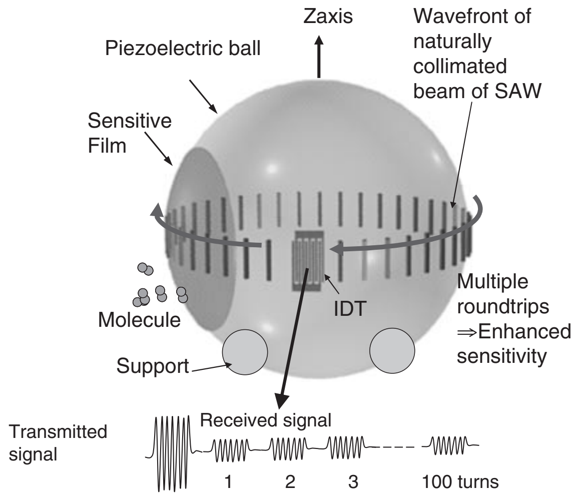
1. INTRODUCTION

Many types of hydrogen gas sensors for leak detection and concentration control have been studied for the safety of fuel cells in coming hydrogen century. For example, field effect transistor (FET), resistivity and surface acoustic wave (SAW) sensors have been proposed [1–5]. However, each sensors have their own drawback in detection range [1], response time [5] or stability [5]. By virtue of the well-developed SAW device technologies [6,7], SAW hydrogen sensor has relatively wide detection range from 0.1% to 100% [5]. However, the response time of SAW sensor is longer than 100 s [5], because relatively thicker sensitive film is needed for SAW sensor. On the other hand, the FET sensor [1] has much shorter response time, since it is so sensitive that very thin sensitive film can be employed. However, the FET sensor is saturated at about 1% H_2, which is a serious drawback since it can not tell us when the lowest explosion limit of 4% is reached.

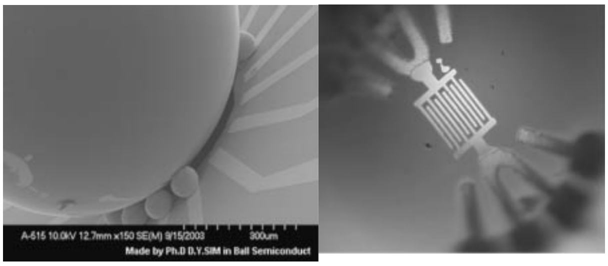
In these circumstances, we have studied property of SAW on a ball and developed ball SAW devices [8–14] based on the multiple roundtrip propagation of SAW along a narrow path on the surface of ball without the diffraction effect [8]. Subsequent theoretical work predicted a naturally collimated beam of SAW [9], which was experimentally verified by using a piezoelectric transducer [10] and a laser probe [11]. Present ball SAW device is composed of a piezoelectric crystal ball with an IDT of a proper width, directly fabricated along a proper route [12–14]. By using a proper length of IDT, collimated SAW beams travel many times along the spherical surface [9–14], and the roundtrip time can be precisely measured [8]. Consequently, we proposed a ball SAW hydrogen sensor [15] with a Pd sensitive film that absorbs hydrogen, causing change of elasticity. The velocity of SAW thus changes as function of hydrogen concentration. By virtue of the excellent sensitivity caused by the multiple roundtrips, we developed a very wide range hydrogen sensor, from 0.001% to 100% [16]. In this paper we describe these topics.

2. SENSORS BASED ON COLLIMATED BEAM OF SURFACE WAVE

The principle of the ball SAW sensor is illustrated in Fig. 1(a). A sensitive film is deposited on the equator of a ball, on which a collimated beam of SAW is generated by an interdigital transducer (IDT) and detected by the same IDT after one, two and many turns. The velocity and attenuation of SAW are changed by the absorption of



(a)



(b)

Fig. 1 Surface acoustic wave sensors. (a) Multiple roundtrips over 100 turns enabling excellent sensitivity. (b) A f 1 mm quartz ball SAW hydrogen gas sensor with 40-mm-thick Pd30%Ni film installed on a glass substrate.

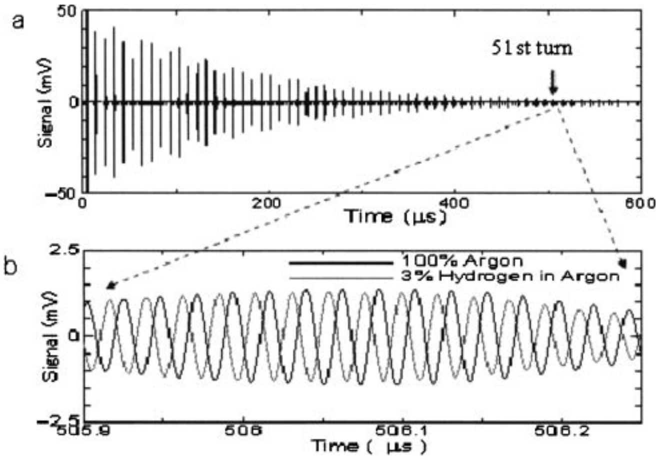


Fig. 2 Waveform of ball SAW sensor (left). (a) 45 MHz SAW signal observed on Z-axis cylinder of f 10 mm quartz ball. (b) Effect of 3% H_2 gas in Ar producing significant difference in delay time at 51st turn.

molecules into the sensitive film. Although the change is small for a very low concentration, it is amplified after ultramultiple roundtrips (more than 50 turns). If the attenuation coefficient variation multiplied by the circumference of the sphere is $\Delta\alpha L = 10^{-3}$, the relative amplitude after one turn is $\exp(-\Delta\alpha L) = \exp(-10^{-3}) = 0.999$, which is practically indistinguishable from unity. However, the relative amplitude in ball SAW sensors after 100 turns is $\exp(-100\Delta\alpha L) = \exp(-0.1) = 0.905$, whose difference from unity can easily be detected.

The advantage of ball SAW is most fully appreciated when applied to a very thin sensitive film, in which the multiple roundtrips enhance the sensitivity of the sensor and the attenuation loss would not cause a serious problem. Though the sensitive film can be deposited to cover the whole propagation path, only part of it may be sufficient, such as shown in Fig. 1(a) for certain applications, so that other areas can be used for other purposes, such as other routes in crystals supporting multiple routes, or removed.

Ball SAW sensor using a sensitive film thinner than ever has a unique advantage in both sensitivity and response time by amplifying the variation in SAW velocity and attenuation of a palladium (Pd) or PdNi film when it adsorbs H_2 molecules [15]. The sensor was fabricated on a φ10 mm single-crystal quartz ball with an acoustic frequency of 45 MHz, and on a φ1 mm single-crystal quartz ball with an acoustic frequency of 156 MHz, and the latter is shown in Fig. 1(b). The IDT was fabricated by photolithography. The Pd or PdNi film was thermally evaporated under a pressure of 10^{-4} Pa at a rate of 0.01 nm/s, with a thickness of 20 nm or 40 nm.

The delay time and amplitude of SAW were measured using an ultrasonic pulser/receiver. Figure 2(a) shows the multiple-roundtrip signal on a φ10 mm device. A SAW with a center frequency of 44.8 MHz was clearly visible after 100 turns, and the total propagation length was 1,601 mm at the 51st turn. Considering the width of the IDT ($2d = 0.7$ mm) and the wavelength of SAW (0.07 mm at 45 MHz), the Fresnel distance is 1.75 mm. Since 1,600 mm is 915 times the Fresnel distance, it is a surprisingly large propagation length that is impossible on a planar or cylindrical surface due to diffraction loss. Since the decay time τ to reach amplitude of 1/e was 200 μs and the frequency f was 45 MHz, the Q factor was evaluated to be $\pi f \tau = 28{,}000$. The performance of the sensor is shown in Fig. 2(b), which is an expanded waveform of Fig. 1(a) at the 51st turn. The waveform observed in 3% H_2/Ar is also plotted here and it is clearly separated from that observed in 100% Ar.

To quantitatively evaluate the delay time of the SAW at 51st turns, the signal was digitized with a sampling frequency of 2 GHz, and the real part of the wavelet transform of the waveform was calculated using the Gabor mother wavelet. Although the sampling period was 0.5 ns, 50 interpolations realized a temporal resolution of 0.01 ns. The noise level was so small after 10 averagings on a

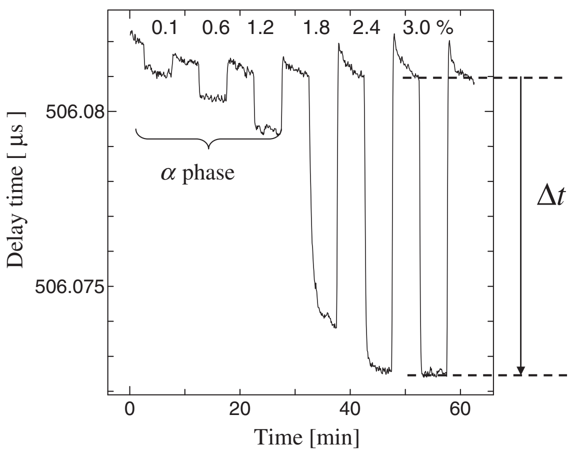


Fig. 3 Ball SAW sensor performance showing delay time response to H_2 (right). H_2 concentration is shown in percentage.

digital oscilloscope, a signal-to-noise ratio of 40 dB was achieved, and a smooth variation in delay time was observed even at the interpolated curve as shown in Fig. 3. Thus, together with the long delay time of 506 μs owing to the large propagation length, the relative resolution of delay time measurement was 0.02 ppm or 2×10^{-8}.

In this resolution regime, a dominant source of error is the temperature dependence of SAW velocity. To eliminate this error, we measured the temperature using a semi-rigid copper-constantan thermocouple pressed against the ball for accurate temperature measurement. Alternatively, thin-film thermometer could also be deposited. The temperature effect compensation was performed using the measured temperature and the temperature coefficient of SAW velocity, 24.6 ppm/° (9.9415 ns/° for a 10-mm-diameter ball after 50 turns) which was previously estimated for the quartz ball.

Other methods for the temperature compensation such as using a reference SAW device without the sensitive film can also be employed. Further development along this approach will be published in the near future.

3. SENSITIVITY OF BALL SAW HYDROGEN SENSOR

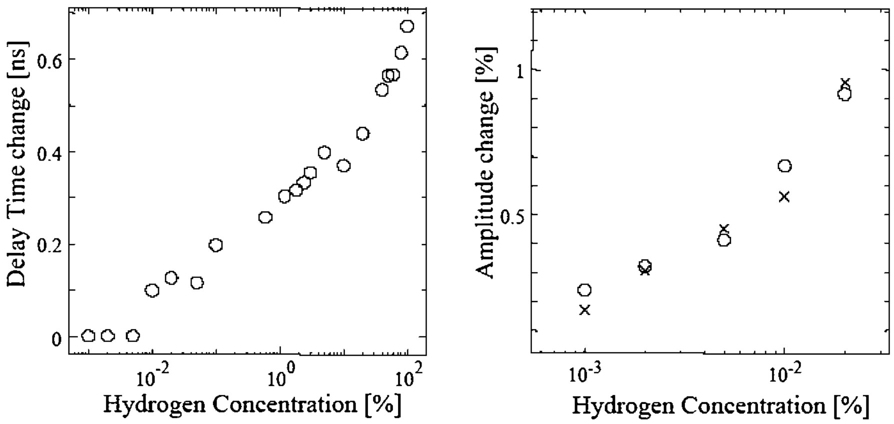
Figure 3 shows a delay time response, the variation in delay time in a measurement sequence for each H_2 concentration [16]. A decrease in delay time was observed due to H_2 and subsequent recovery was observed due to 100% Ar. Even at the lowest concentration of 0.1% H_2, a delay time response of 0.5 ns (1 ppm in 506 μs) was clearly observed. Since the variation is much larger than the noise, these data potentially show a sensitivity to lower concentrations. Below 1.2% H_2, the response time was 20 s for full scale change. In a recent study, the response time was as fast as 2 s and it was 1/50 of the response time of previous fastest SAW H_2 sensor operated at room temperature [5]. Such a difference in response time is probably caused by a smaller film thickness (20 nm) of ball SAW sensor compared with the previous one (100 nm) [5].

From the delay time difference and total propagation length, the SAW velocity increase in 3% H_2 was evaluated to be 0.054 m/s. This increase can be explained by theoretical calculation based on the transfer matrix approach. We calculated the SAW velocity increase due to the decrease in density ρ, and increase in longitudinal modulus c_{11} or shear modulus c_{44}, independently. A 20-nm-thick isotropic Pd film and single-crystal quartz at 45 MHz were assumed and the calculated velocity was averaged for different cuts along the z-cylinder at 10° intervals. We found that a 1% decrease in ρ, a 3% increase in c_{11} or a 40% increase in c_{44} can independently account for the velocity increase of 0.054 m/s at 45 MHz. This change is consistent with reported changes in elasticity of Pd in hydrogen.

We now evaluate the variation in SAW attenuation using the amplitude response of a φ1 mm quartz ball SAW sensor operated at 156 MHz. The φ1 mm sensor was installed in a package with an IDT fabricated on the bottom of the ball as shown in Fig. 1(b). A 40-nm-thick PdNi film [1] was deposited on top of the ball with a 25% coverage of the whole equator path. Roundtrips of SAW consisting of as many as 200 turns were observed, but the signal at the 50th turn was used to obtain an amplitude response.

Much lower H_2 concentration of 0.05% (500 ppm) and 0.001% (10 ppm) were detected from the amplitude response, as shown in Fig. 4. We found a clear response even at the lowest H_2 concentration of 10 ppm, with a signal-to-noise ratio of four. This is the highest sensitivity thus far reported with SAW H_2 sensors. The higher H_2 concentration of 0.01% (100 ppm) and 100% were detected from the delay time response, as shown in Fig. 5. We found a change in delay time response Δt (defined in Fig. 2) even at the highest H_2 concentration of 100% without saturation [16].

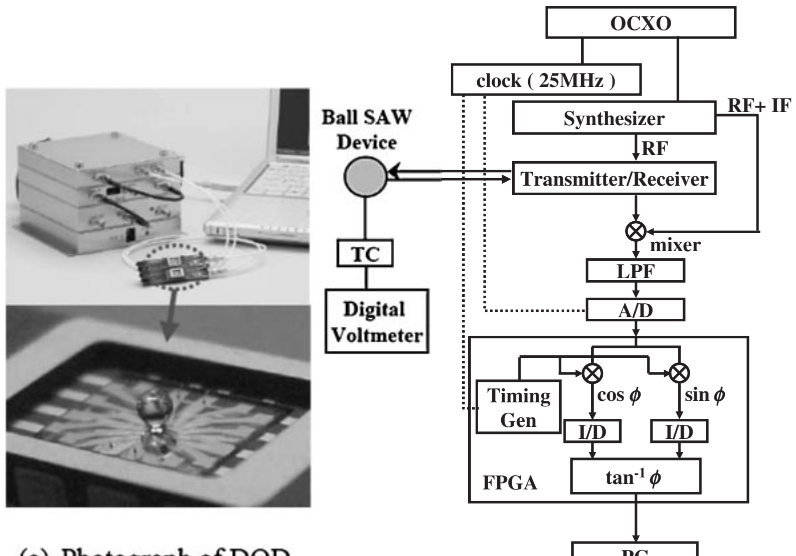
However, the evaluation of response time was not satisfactory since the measurement interval was relatively long (2 s) due to the need for averaging of signals (up to 20 times) in the digital oscilloscope [15,16]. In order to reduce the measurement intervals, we developed a digital quadrature detector (DQD) using a field programmable gate array (FPGA) shown in Fig. 5 [17]. The transmitter and receiver of a tone burst RF signal were connected to the ball SAW sensor, and the SAW signal after multiple roundtrips was detected by the heterodyne detection. The phase φ was calculated from the cos φ and sin φ obtained by the quadrature demodulation [17]. The phase data were averaged inside the integration and dump (I/D) filter in the FPGA at a fast rate of 1 kHz, and 256 averaging was achieved only in 0.256 s.



(a) Delay time change (in high concentration)

(b) Amplitude change (in low concentration)

Fig. 4 Hydrogen detection to the range of 10 ppm to 100% concentration.



(a) Photograph of DQD with ball SAW hydrogen sensor

(b) System block diagram

Fig. 5 High speed measurement system of ball SAW hydrogen sensor.

Figure 6 shows delay time responses to hydrogen gas showing better S/N ratio by virtue of the DQD. The response time was found to be less than 1 s at 3% hydrogen concentration in (b), since only four points were observed in the transition indicated by the arrow.

Although the FET [1] and tunnel diode [2] H_2 sensors have similar sensitivities to that of the ball SAW sensor, they have a drawback of saturation in the critical H_2 concentration range of roughly 1%. Since the PdNi film does not suffer from the degradation by phase transition and maintains stability even after repeated exposures to 100% H_2 [1], and the present ball SAW sensor does not saturate up to 100%, it would be feasible to develop a ball SAW H_2 sensor using a PdNi film that covers a wide concentration range of 10 ppm to 100%. This is the first single-element H_2 sensor fabricated to date, and will satisfy the sensor needs for fuel cell systems and direct-injection engines.

4. CONCLUSION

The ball SAW hydrogen sensor achieved the widest sensing range of 10 ppm to 100% for the first time. We have verified the response time less than 1 s to 3% hydrogen gas using a newly developed DQD, which is ready for application to sensors of many other gases or odors.

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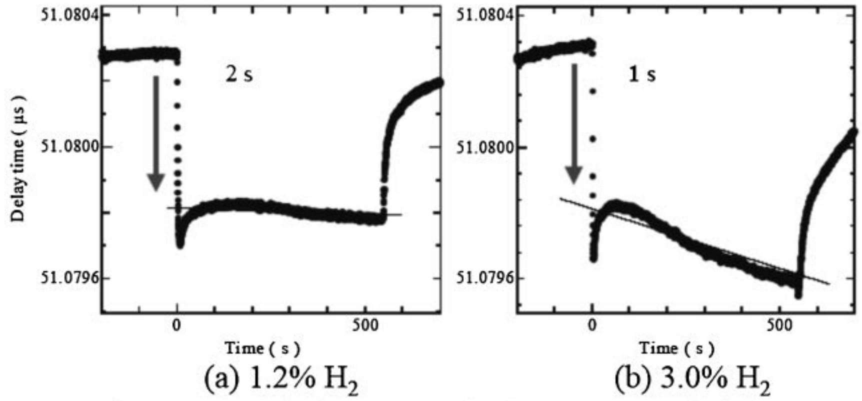

Response time ≤ 1s, for 3% hydrogen ; Averaging time : 256, Interval of points : 0.256 s

Fig. 6 Evaluation of response time using DQD.

T. Mihara, A. Mizukami and T. Tsuji (Tohoku University), N. Nakaso, S. Akao, T. Ohgi, T. Nakatsukasa and Y. Tsukahara (Toppan Printing Co. Ltd.), D.-Y. Sim, B. Maxey and N. Takeda (Ball Semiconductor Inc.) and I. Satoh, H. Kazato, T. Fukiura, H. Tanaka, T. Miyagishi and Y. Ebi (Yamatake Corp.).

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