RESEARCH ARTICLE

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Sensitive Capacitive-type Hydrogen Sensor Based on Ni Thin Film in Different Hydrogen Concentrations

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Abstract: Background: Hydrogen sensors are micro/nano-structure that are used to locate hydrogen leaks. They are considered to have fast response/recovery time and long lifetime as compared to conventional gas sensors. In this paper, fabrication of sensitive capacitive-type hydrogen gas sensor based on Ni thin film has been investigated. The C-V curves of the sensor in different hydrogen concentrations have been reported.

Method: Dry oxidation was done in thermal chemical vapor deposition furnace (TCVD). For oxidation time of 5 min, the oxide thickness was 15 nm and for oxidation time 10 min, it was 20 nm. The Ni thin film as a catalytic metal was deposited on the oxide film using electron gun deposition. Two MOS sensors were compared with different oxide film thickness and different hydrogen concentrations.

Results: The highest response of the two MOS sensors with 15 nm and 20 nm oxide film thickness in 4% hydrogen concentration was 87.5% and 65.4% respectively. The fast response times for MOS sensors with 15 nm and 20 nm oxide film thickness in 4% hydrogen concentration was 8 s and 21 s, respectively.

Conclusion: By increasing the hydrogen concentration from 1% to 4%, the response time for MOS sensor (20nm oxide thickness), was decreased from 28s to 21s. The recovery time was inversely increased from 237s to 360s. The experimental results showed that the MOS sensor based on Ni thin film had a quick response and a high sensitivity.

Keywords: Thin film, capacitive, oxide thickness, recovery, sensor, concentration.

1. INTRODUCTION

Hydrogen sensors generate an electrical signal that is proportional to hydrogen concentration in air. Hydrogen sensors are micro/nano-structure that are used to locate hydrogen leaks. They are considered to have fast response/recovery time and long lifetime as compared with conventional gas sensors. The first report of hydrogen gas sensor based on metal-oxide-semiconductor (MOS) structure was reported in Ref. [1, 2]. The MOS sensor consists of triple layers of metal/oxide/semiconductor. Hydrogen molecules on the catalytic metal surface dissociated to atomic hydrogen. Hydrogen atoms diffuse into metal and are absorbed at the metal/oxide interface and polarized. The dipole layer in the interface changes the work function of the metal. Palladium (Pd), nickel (Ni) and platinum (Pt) are used in many of hydrogen gas sensors with two forms, nanoparticles and thin film. Pd nanoparticles (PdNPs) resistive-type hydrogen gas sensor at 50°C temperature and in 0.1-1% hydrogen concentration showed the response time 10 s and recovery time 31 s

[3]. Composite of Pd nanoparticles with carbon nanotubes (CNTs) showed fast response time for resistive-type hydrogen gas sensors [4-7]. For nanocomposite CNTs/Ni-PdNPs at room temperature and 0.2-5% hydrogen concentration the response and recovery times were 20 s and 140 s respectively [8]. Comparison of Pd nanoparticles/graphene hydrogen gas sensors indicated that at room temperature the response times of these sensors were faster than recovery times [9-12]. Phan *et al.* [13] highlighted that for nanocomposite graphene/Ni-PdNPs at room temperature and 0.001-0.1% hydrogen concentration the response time was 180 s and the recovery time was 720 s. Palladium thin film was used in many of hydrogen gas sensor with different sensing physical parameter such as resistance, current and voltage [14-20]. Chung *et al.* [21] demonstrated that at 150°C and 1% hydrogen concentration, the response and recovery times for nanocomposite Pt/Pd thin film were 4s and 5s respectively. In Ref [22], a capacitive-type hydrogen sensor was fabricated with the Ni thin film. These authors showed at 150°C and in 0-1% hydrogen concentration of 14.8 nm oxide thickness, the response time was 875 s and the recovery time was 45 min. In this paper fabrication of sensitive capacitive-type hydrogen gas sensor based on Ni thin film has been investigated. The

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C-V curves of the sensor in 0%, 1%, 2%, 3% and 4% hydrogen concentration at 120°C have been reported. Moreover, the response/recovery time of the sensor with different hydrogen concentration were compared.

2. MATERIALS AND METHOD

A capacitive-type hydrogen gas sensor with the structure of Ni/SiO₂/Si has been fabricated. The overall process flow of the MOS sensor has been illustrated in Fig. (1). The semiconductor is <400> n-type silicon (0.22 Ω cm). Dry oxidation was done in thermal chemical vapor deposition furnace (TCVD). In presence of pure nitrogen gas the TCVD furnace ramp up to 1000°C. In 1000°C the nitrogen gas was turned off and O₂ gas with 2 lit/min was opened. For oxidation time 5 min the oxide thickness was 15 nm and for oxidation time 10 min, it was 20 nm. The surface of the oxide film was characterized by AFM module (DS 95-50-E scanner). The AFM images of the oxide film have been depicted in Fig. (2). The average roughness for the oxide film thicknesses 15 nm and 20 nm were 10.5 nm and 3 nm respectively. The summits and valleys of AFM image in same directions for the oxide film thicknesses 15 nm and 20 nm have been shown in Fig. (3). As can be seen in Fig. (3), the highest summits (peak height above the brain profile) for oxide film thicknesses 15 nm and 20 nm were 100 nm and 20 nm respectively. The effects of oxide film thickness and surface texture parameters on the performance of the MOS sensor are considerable. On the oxide film a 100 nm film of Ni thin film was deposited using electron gun deposition. As an ohmic contact, an Au film with 100 nm thickness was deposited on the backside of the Si using evaporating. The electrodes were connected to the gate and Au film. For capacitance measurement of the MOS the wires were attached to the LCR meter. The set up includes LCR meter modules (GPS-3135B) that can be interfaced to a PC. The LCR meter is capable of measuring precisely capacitance values in the frequency range 50 Hz to 100 KHz.

3. RESULTS AND DISCUSSION

The flat-band voltage (V_{FB}) is the boundary between accumulation and depletion layers. The V_{FB} is given by [23]:

$$V_{FB} = \frac{W_M - W_S}{q} - \left(\frac{Q}{C_{ox}} \right) \quad (1)$$

Where W_M and W_S are the work function of gate metal and semiconductor and Q is the trapped charges in the oxide film. The trapped charges have four sources: (i) Fixed oxide charges (Q_f) (ii) Mobile ionic charge (Q_m) (iii) Interface trapped charges (Q_{it}) (iv) Oxide trapped charges (Q_{ot}) [24-28]. The work function of Ni varies from 5 eV to 5.35 eV for Si varies from 4.6 eV to 4.8 eV. The typical value of Q For a n-type MOS sensor is 3.2×10^{-9} (C cm⁻²) [26]. The energy band diagram of ideal MOS sensor in unbiased condition is shown in Fig. (4). The diagram shows the different energy levels for MOS structure. The Fermi levels in the metal gate and semiconductor are indicated by E_{FM} and E_{FS} respectively. The χ_s is the electron affinity in the semiconductor. As can be seen in Fig. (4), the work function of metal is equal to the work function of semiconductor and all energy

Fig. (1). Overall process flow of the MOS sensor.

Fig. (2). AFM images of the oxide film thickness (a) 15 nm (b) 20 nm.

Fig. (3). Summits and valleys of AFM image in same directions for the oxide film thicknesses 15 nm and 20 nm.

Fig. (4). Energy band diagram of ideal MOS sensor in unbiased condition.

level are flat. Also, Fig. (4) shows the trapped charges in the oxide film and at the oxide/semiconductor interface are zero. Therefore for an ideal MOS sensor, the V_{FB} is zero. The energy band diagram of real MOS sensor in unbiased situation is shown in Fig. (5). In the real MOS sensor, the work functions of metal and semiconductor are different. Fig. (5) shows the energy barriers between metal/oxide and oxide/semiconductor. The importance of energy barriers is that they prevent the free flow of carriers from metal to semiconductor. For the real Ni/SiO₂/Si sensor the V_{FB} is 0.2 V. When a positive voltage is applied to the MOS sensor, electrons are attracted at the oxide/semiconductor interface and create an accumulation layer. When the MOS sensor is biased by negative voltage, the voltage repels electrons and creates a depletion layer. By increasing the negative voltage of the MOS capacitor sensor, the n-type semiconductor enters an inversion layer. The capacitance of depletion layer is obtained from [23]:

Fig. (5). Energy band diagram of real MOS sensor in unbiased situation.

$$C = \frac{\epsilon_{ox}\epsilon_0 A}{t_{ox} + \left(\frac{\epsilon_{ox}}{\epsilon_s}\right)X_d} \quad (2)$$

Where ϵ_{ox} is the relative permittivity of the oxide film, A is the area of the Pd gate, t_{ox} is the thickness of the oxide film, ϵ_s is the relative permittivity of the Si and X_d is the width of the depletion layer. For the 15 nm oxide thickness the capacitance of MOS sensor in the accumulation layer is 230 nF and for 20 nm oxide, the thickness is 172 nF. The capacitance of MOS sensor before the inversion layer for 15 nm and 20 nm oxide thicknesses are 30 nF and 29 nF respectively. The capacitance-voltage (C-V) curves of the MOS sensor with 15 nm oxide thickness at the 120°C temperature and 0%, 1%, 2%, 3% and 4% hydrogen concentration are shown in Fig. (6). As can be seen in Fig. (6), all curves have the same trend and the capacitance falls to the negative voltage side. By increasing the hydrogen concentration, the capacitance of the MOS sensor in depletion layer is increased. Fig. (6) shows the experimental capacitance of the MOS sensor is smaller than the theoretical value. This behavior is related to the relative permittivity of the oxide film. The relative permittivity of the oxide film is complex. The real and imaginary parts of the relative permittivity depend on the measurement frequency. In the MOS sensor by increasing the measurement frequency, the real part of the relative permittivity is decreased [27]. This behavior is related to the fact that by increasing the frequency, the interfacial dipoles have a smaller time constant for orienting themselves in the direction of the field. For a MOS sensor, by increasing the frequency from 1 KHz to 100 KHz, the relative complex permittivity decreases from 3.9 to 2.75. Fig. (6) shows that the experimental value of the V_{FB} in 0% hydrogen concentration and in pure nitrogen is 0.5 V. The difference between experimental and theoretical values of V_{FB} is related to the trapped charges in the oxide film [23]. The standard technique to measure the oxide trapped charges is C-V measurement. From Eq. 1 for MOS sensor with 15 nm oxide thickness, the flat-band voltage shift is 0.3 V and the number

Fig. (6). C-V curves of the MOS sensor with 15 nm oxide thickness at the 120°C temperature and 0%, 1%, 2%, 3% and 4% hydrogen concentration.

of the oxide trapped charges is increased from 3.2×10^{-9} to 51.8×10^{-9} (C cm^{-2}). The C-V curves of the MOS sensor with 15 nm oxide thickness have been shown in Fig. (6) and the C-V curves of the MOS sensor with 20 nm oxide thickness have been shown in Fig. (7) but for better comparison, the C-V curves of the MOS sensor with 15 nm oxide thickness as shown in Fig. (7) (the C-V curves of the MOS sensor with 15 nm oxide thickness have been repeated in Fig. (7)). Comparison of C-V curves for different oxide thicknesses shows that by increase the oxide thickness from 15 nm to 20 nm the capacitance of the MOS sensor in accumulation layer is decreased from 162 nF to 121 nF. Fig. (7) also shows that the experimental value of the V_{FB} in 0% hydrogen concentration and in pure nitrogen is 0.7 V. From Eq. 1 for MOS sensor with 20 nm oxide thickness, the flat-band voltage shift is 0.5 V and The number of the oxide trapped charges is increased from 3.2×10^{-9} to 51.8×10^{-9} (C cm^{-2}). Comparison of C-V curves for MOS sensors when exposed to hydrogen gas shows that flat-band voltages were decreased. The mechanism of hydrogen detection is that the hydrogen atoms at the metal/oxide interface create a dipole layer. This dipole layer causes a decrease in the metal work function. The response of the MOS sensors is obtained from:

$$R(\%) = \left(\frac{C_H}{C_N} - 1 \right) \times 100 \quad (3)$$

Where C_N is the capacitance in 0% hydrogen concentration and C_H is the capacitance in a 1%, 2%, 3% and 4% hydrogen concentration. The response ($R\%$) of the MOS sensor for 15 nm oxide thickness is shown in Fig. (8). The response ($R\%$) of sensor to 1%, 2%, 3% and 4% hydrogen concentration is 60%, 72.5%, 80% and 87.5% respectively. Fig. (8) shows that the response ($R\%$) of the MOS sensor increased by rising the hydrogen concentration. By rising the hydrogen concentration, the number of hydrogen atoms at the interface is increased. Therefore, the dipole layer in the metal/oxide

Fig. (7). Comparison of C-V curves of the MOS sensors with 20 nm and 15 nm oxide thicknesses at 120°C temperature and 0%, 1%, 2%, 3% and 4% hydrogen concentration.

Fig. (8). Response ($R\%$) of the MOS sensor for 15 nm oxide thickness.

Fig. (9). Response ($R\%$) of the MOS sensor for 20 nm oxide thickness.

interface becomes stronger. The response (R%) of the MOS sensor for 20 nm oxide thickness is shown in Fig. (9). The response (R%) of sensor to 1%, 2%, 3% and 4% hydrogen concentration is 43.2%, 53%, 58% and 65.4% respectively. Comparison of Fig. (8) and Fig. (9) shows that the highest response (R%) occurs in the voltage 0.14 V. Also, the response (R%) of the MOS sensor with thin oxide film is higher than thicker oxide film. The main reason for this behavior is related to the trap states at the metal/oxide interface. The trap states for thinner oxide film are much greater than that of the thicker oxide film, which tends to be a stronger dipole layer at metal/oxide interface. The physical mechanism of the association of trap states is shown in Fig. (10). The trap states in the oxide film as denoted by the bubbles. The thicknesses of the oxide films are shown by dash lines. It can be seen in Fig. (10) the density of trap states decreased by increasing in the oxide film thickness. This mechanism is similar to the one proposed earlier [22]. The response time ($t_{90\%}$) is the time for reaching 90% of the signal magnitude and the recovery time ($t_{10\%}$) is the time interval for recovery of 10% of the signal magnitude. The response/recovery time of MOS sensor with 15 nm oxide film thickness to 1%, 2%, 3% and 4% hydrogen concentration in voltage 0.14 V and at 120°C is shown in Fig. (11). The response times for 1%, 2%, 3% and 4% hydrogen concentration were 12 s, 10.6 s, 9 s and 8 s respectively. The recovery times for 1%, 2%, 3% and 4% hydrogen concentration were 113 s, 161 s, 206 s and 240 s respectively. Fig. (11) shows that by rising the hydrogen concentration, the response time is decreased. This behavior is related to the decrease of the metal work functions and a larger shift in the V_{FB} . Comparison of response/recovery time of MOS sensor to different hydrogen concentration is shown in Table 1. As can be seen in Table 1 for the MOS sensor with 20 nm oxide thickness by increase in the hydrogen concentration from 1% to 4%, the response times are decreased from 28 s to 21 s. As a result from Fig. (10) and Fig. (11), the number of hydrogen atoms in the Ni/SiO₂ interface increases with the decrease in the oxide film thickness. The sensing mechanism of the MOS sensor is that of the association of trap states in the Ni/SiO₂ interface. The increase of trap states is caused a stronger dipole layer at the Ni/SiO₂ interface and therefore, the metal work function is reduced. The comparison of the performance of hydrogen sensors based on Ni thin film is shown in Table 2. It can be

Fig. (10). Physical mechanism of the association of trap states.

Fig. (11). Response/recovery time of MOS sensor with 15 nm oxide film thickness to 1%, 2%, 3% and 4% hydrogen concentration in voltage 0.14 V and at 120°C temperature.

seen in Table 2 that for the operating temperature 120-150°C, the response time of Ni thin film hydrogen gas sensors [22, 28, 29] is of the order of few seconds and the recovery time is of the order of few minutes. Table 2 shows that combination of Ni thin film with Pd by different sensing parameters, the operating temperature of the sensors is eliminated and also the response time of the sensors is reduced [30-33]. For NiO thin film/Pd nanoparticles at temperature range 53-180°C, the response time is 3 minutes [34]. Aguilar [35] shows for single wall carbon nanotubes (SWCNTs) loaded with Ni-Pd nanoparticles for hydrogen detection at room temperature that the response time and recovery time are 20 s and 140 s, respectively. For Ni-Pd/graphene thin film hydrogen sensor at room temperature and 0.1% hydrogen concentration, the response time is 3 minutes and recovery time is 12 minutes [13]. Table 2 shows for the Ni-Mg thin film hydrogen sensor in 0.001-10% hydrogen concentration, the response time and recovery time are 50 s and 100 s, respectively [36].

CONCLUSION

In this paper, fabrication sensitive capacitive-type hydrogen gas sensor based on Ni thin film has been investigated. The 15 nm and 20 nm oxide film thicknesses were created using dry oxidation and analyzed by AFM. Comparison of C-V curves for different oxide thicknesses shows that by increase in the oxide thickness from 15 nm to 20 nm, the capacitance of the MOS sensor in accumulation layer is decreased from 162 nF to 121 nF. The response (R%) of sensor with 15 nm oxide thickness to 1%, 2%, 3% and 4% hydrogen concentration was 60%, 72.5%, 80% and 87.5% respectively. For the MOS sensor with 20 nm oxide film thickness, the response (R%) to 1%, 2%, 3% and 4% hydrogen concentration was 43.2%, 53%, 58% and 65.4% respectively. The response times for sensor with 15 nm oxide film thickness to 1%, 2%, 3% and 4% hydrogen concentration were 12 s, 10.6

Table 1. Comparison of response/recovery time of MOS sensors in different hydrogen concentrations.

Oxide Film Thickness	Detection Limit	Response Time ($t_{90\%}$)	Recovery Time ($t_{10\%}$)
15 nm	1%	12 s	113 s
	2%	10.6 s	161 s
	3%	9 s	206 s
	4%	8 s	240 s
20 nm	1%	28 s	237 s
	2%	25 s	291 s
	3%	23.4 s	318 s
	4%	21 s	360 s

Table 2. Comparison of the performance of the hydrogen sensors based on Ni thin film.

Material	Temperature Range (°C)	Detection Limit (%)	Sensing Parameter	Response Time	Recovery Time	References
Ni thin film	120	1-4	Capacitance	8 s	240 s	This work
Ni thin film	150	1-4	Capacitance	16 s	680 s	[22]
Ni thin film	140	0.05-0.1	Capacitance	44 s	630 s	[28]
Ni thin film	140	4	Capacitance	84 s	45 min	[29]
Ni-Pd nanoparticles film	Room temperature	0-5 KPa	Conductance	<1 s	-	[30]
Ni-Pd thin film	Room temperature	0.01-10	Resistance	5 s	-	[31]
Ni-Pd thin film	Room temperature	0-10	Current	<1 s	-	[32]
Ni-Pd thin film	Room temperature	0-4	Wavelength	4-5 min	-	[33]
NiO thin film- PdNPs	53-180	0.1-1	Resistance	3 min	-	[34]
Ni-Pd nanoparticles film-SWCNTs	Room temperature	0.2-5	Resistance	20 s	140 s	[35]
Ni-Pd/ graphene thin film	Room temperature	0.0001-0.1	Resistance	3 min	12 min	[13]
Ni-Mg thin film	Room temperature	0.001-10	Resistance	50 s	100 s	[36]

s, 9 s and 8 s respectively. For the MOS sensor with 15 nm oxide film thickness, the recovery times for 1%, 2%, 3% and 4% hydrogen concentration were 113 s, 161 s, 206 s and 240 s respectively. In the MOS sensor with 20 nm oxide thickness, with increase in the hydrogen concentration from 1% to 4%, the response times decreased from 28 s to 21 s and the recovery times increased from 237 s to 360 s. The capacitive-type hydrogen gas sensor shows very fast response and high sensitive.

ETHICS APPROVAL AND CONSENT TO PARTICIPATE

Not applicable.

HUMAN AND ANIMAL RIGHTS

No Animals/Humans were used for studies that are base of this research.

CONSENT FOR PUBLICATION

Not applicable.

CONFLICT OF INTEREST

The authors declare no conflict of interest, financial or otherwise.

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