졸업논문청구논문

The Characteristic of the 2 Dimensional Electron Gas based Flexible Hydrogen Sensor manufactured by Atomic Layer Deposition

ALD 공법으로 제작한 2DEG 기반의 유연한 수소 센서의 특성

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A thesis submitted to the Gyeonggi Science High School in partial fulfillment of the requirements for the graduation. The study was conducted in accordance with Code of Research Ethics[1].

2019. 6. 18.

Approved by

Teacher Ha, Sang-woo

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The Characteristic of the 2 Dimensional Electron Gas based Flexible Hydrogen Sensor manufactured by Atomic Layer Deposition

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Abstract

The current hydrogen sensors have various difficulties such as being able to measure only at a high temperature and high concentration. On the other hand, hydrogen sensors based on 2DEG have been reported to perform well at room temperature. This paper focuses on constructing the hydrogen sensor using Al_2O_3/STO 2DEG manufactured by ALD method on the flexible Polyimide plate. Through the bending experiment, the manufactured hydrogen sensor's performance has been measured. The response time, recovery time and sensitivity did not show any significant change before and after bending. Also, compared to currently existing hydrogen sensors, the sensor in this paper showed a huge increase in its performance. The results in the paper shows that the manufactured flexible hydrogen sensor could be used in different situations, showing resistance to physical stress.

ALD 공법으로 제작한 2DEG 기반의 유연한 수소 센서의 특성

초 록

현재 수소 센서는 고온 및 고농도에서만 측정 할 수 있는 등의 여러 가지 어려움이 있다. 반면 2DEG 기반의 수소 센서는 실온에서 잘 작동하는 것으로 보고되었다. 본 논문에서는 ALD 방식으로 제조 된 Al_2O_3/STO 2DEG를 공정 과정에서 변형이 될 수 없는 Polyimide 판에 수소 센서를 구성하여 수소 센서를 구부릴 수 있도록 만드는데 초점을 맞추고 있다. 본 연구에서 제작된 수소 센서의 response time, recovery time 및 sensitivity는 구부리기 전과 후에 큰 변화를 보이지 않았다. 또한 기존의 수소 센서에 비해 센서의 성능이 크게 향상되었음이 보고되었다. 따라서 본 연구의 결과를 통해 ALD 방식으로 제조 된 유연한 수소 센서가 많은 상황에서 사용될 수 있음을 보일 수 있었다.

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Ⅰ. Introduction

As the supply of fossil fuels decreases, the importance of environmentally friendly energy grows. One of the fastest growing fields is in hydrogen energy. Among many hydrogen energy-based technologies, the hydrogen-fueled car is a major technology, as many car companies are aiming for the commercialization of hydrogen-fueled cars. However, there are also some dangers of hydrogen-fueled cars. One of the main challenges of developing a hydrogen-fueled car is to stabilize the hydrogen to prevent the ignition or explosion of the gas. In order to use hydrogen energy safely, it is significant to know the leakage of hydrogen. This is when an accurate hydrogen sensor is needed, to check the concentrations of the hydrogen every second. Hydrogen sensors are mainly classified into 8 different types; Catalytic, Thermal conductivity, Electrochemical, Resistive, Work function, Mechanical, Optical, or Acoustic types, based on their operating methods [1]. The majority of the sensors' distinguishable concentration is 5~10%, which is very large. The sensors also have a problem because their usable temperature is too high compared to the room temperature.

Although recent studies have shown the possibility of a 2DEG hydrogen sensor, the current hydrogen sensor is not flexible [2]. 2DEG is the abbreviation of 2 dimensional electron gas, which is a layer that electrons can move in. Specific details will be covered in the section entitled Theoretical Background. The fact that the hydrogen sensor is not flexible makes it hard to for hydrogen sensors to be

attached to different places. Also, an inflexible sensor shows weakness to external forces and change, so a flexible sensor is needed in order to solve these problems. Therefore, this study will focus on making a flexible hydrogen sensor. The goal of this study is to quantitatively and precisely estimate the usability of flexible hydrogen sensors when commercialized in the future through the practical fabrication of flexible hydrogen sensors. In order to satisfy the bending physical conditions, the sensor fabrication process should be limited to a lightweight and miniaturized form as a whole, and the proposal of the process is suggested. Therefore, this study is proposing a hydrogen sensor that can satisfy physical flexibility among various kinds of hydrogen sensors. Further more, an additional process for adding flexibility in the manufacturing process while taking advantage of the principle of existing hydrogen sensors has been explained. Particularly, since the part where the flexible sensor is used must function continuously in the field using the actual hydrogen power, the type of the sensor is limited to the reusable sensor instead of the disposable one.

In order to clarify the practicality of the manufactured sensor, the first step of the study is to measure the performance of the steady state hydrogen sensor compared to the performance of the sensor of a similar condition which is commonly used after the definite sensor fabrication process. The second step is to quantitatively compare the performance change after the actual physical stress is applied to evaluate practicality, since the sensor to be manufactured is aimed at a reusable sensor rather than one-time use. This paper demonstrates the process of making a flexible hydrogen sensor. The sensor is manufactured by the following methods. First, the

ALD is used to fabricate the 2DEG structure on a STO layer. After forming a 2DEG, the E-Beam evaporator is used to deposit the Pd on top of the 2DEG. After this process, In is injected in order to measure the efficiency of the sensor. This paper also shows the process of measuring the electric current depending on the hydrogen concentration. The data of the hydrogen concentration each second obtained by the manufactured sensor is used to calculate the response time, recovery time and the sensitivity. The data in this paper shows that a flexible hydrogen sensor could be made by the steps mentioned above, and additionally showed that the efficiency of the sensor was positive compared to current hydrogen sensors [3].

Ⅱ. Theoretical Background

Ⅱ.1. ALD

The majority of the sensors are made by vapor deposition. Among these depositions, there are mainly 2 ways to construct a nano-scale film. The first method is Physical Vapor Deposition(PVD), and the second method is Chemical Vapor Deposition(CVD). PVD is divided into Physical Layer Deposition(PLD), Thermal expansion method and sputtering method. Chemical Vapor Deposition is divided into thermal CVD and plasma enhanced CVD. Most CVD is LPCVD or UHVCVD.

Atomic Layer Deposition(ALD) method is a type of vapor deposition, specifically Chemical Vapor Deposition (CVD). ALD is made by repeating the process of continuously injecting the reactive gas into the substrate and continuously stacking the layers. The advantage of ALD is that the growth rate is constant. In other words, it builds up exactly one layer of atoms at a time. This makes the sensor's characteristic more constant. Also, the layers could be deposited by a desirable thickness. Substrates that can be used for the ALD process include various types of metals, such as Al_2O_3, SiO_2, oxide and silicon. The deposition process through the ALD method is as follows. First, when trimethylaluminum (TMA) is injected, the substrate is adsorbed to form the first layer. Subsequently, a process called purge is conducted. This process is performed by blowing an inactive gas such as N2 or Ar to remove the remaining source and products in the air. After the purge, water is injected. The water reacts with the material absorbed on the substrate, which results in a deposited layer. This process can be repeated to deposit the material to a

desired thickness in Angstrom units. Therefore, ALD can be deposited in the form of a thin film.

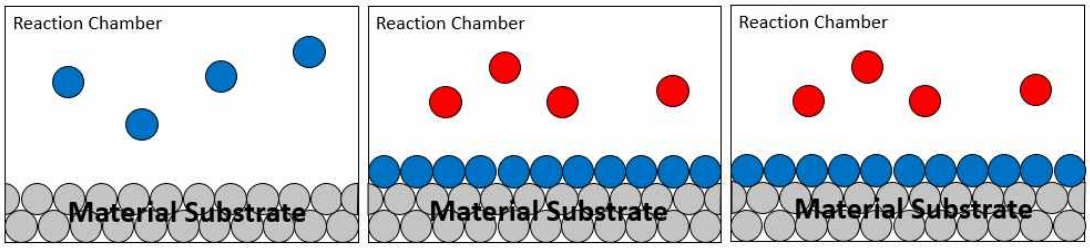

Figure 1. Qualitative process of the ALD method The figure shows the process of the Atomic Layer Deposition. The process of continuously injecting the reactive gas and purging the gas results in stacking the layers. [4]

Ⅱ.2. E-Beam Evaporator

E-Beam Evaporator is a type of ion beam evaporator that deposits materials using electrons as ions. During the sputtering process, the high energy electron beam can actively induce the movement of the evaporation source in a vacuum [5]. Previous research has shown that an E-Beam Evaporator can produce epitaxial growth at the Pd/Ni interface [6]. In this study, the E-Beam Evaporator is used to deposit Pd, which is known to be a nanoparticle. The E-Beam Evaporator is used because it could increase the surface area of the Pd. Pd was used because Pd is known to react well to hydrogen.

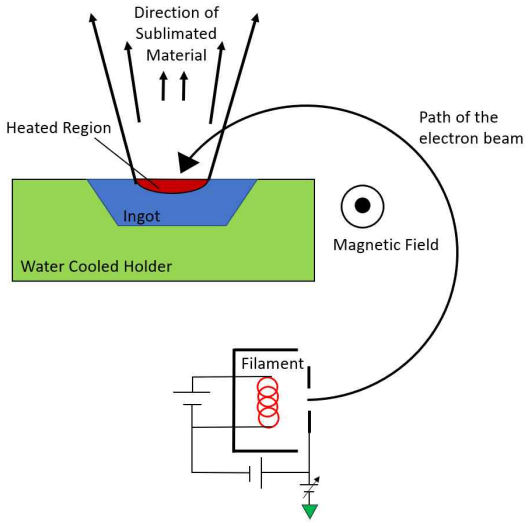

Figure 2. Principle of E-Beam Evaporator The figure shows the principle of E-Beam Evaporator. The high energy elctron can induce the movement of the evaporation source in a vacuum [7]

Ⅱ.3. 2DEG (2-Dimensional Electron Gas)

2-Dimensional Electron Gas(2DEG) is a 2-dimensional electron-layer. The electrons can move freely on the 2-dimensional plane, but their location is limited within the small vertical area. An oxide-based 2DEG is made by the heterojunction of two different oxides. When two different layers of crystalline semiconductors meet and create an interface, it is called a heterojunction. The most well known Oxide-based 2DEG is formed by growing epitaxial $LaAlO_3$(LAO) on a single crystal $SrTiO_3$(STO). This process is performed by a technology called Pulsed Laser Deposition(PLD) at a high temperature of 700~800°C. It is an advanced and expensive technique, and the 2DEG's structure makes it hard to commercialize. However, it has been reported that by the ALD method of construction, an Oxide-based 2DEG could be developed, making it possible to reduce the current process [8].

2DEG hydrogen sensors generally measure the hydrogen concentration by measuring the change in the amount of current due to the change in the number of electrons in the 2DEG. This variation in the number of electrons is mostly induced by the Pd layer deposited on the np diode in 2DEG form. Therefore, the materials forming the 2DEG can be variously presented. In fact, in previous studies, hydrogen sensors have been fabricated through various heterojunctions such as LAO/STO 2DEG, oxide/AlGaAs 2DEG, and AlGaN/GaN 2DEG [2]. Since the 2DEG sensor only requires n-p diode fabrication in 2DEG and Pd deposition, the process can be varied. One of them, the 2DEG hydrogen sensor, can be made by the ALD process and E-Beam Evaporator. When 2DEG is manufactured, 2DEGs of the type represented by LAO / STO 2DEG can be deposited by the ALD method. The Pd layer is also physically deposited on the 2DEG using an e-beam evaporator. Finally, In was injected through Pd-Al_2O_3-2DEG-$SrTiO_3$. The In is used to connect the 2DEG to the conductor to measure the change of the electron number in the 2DEG.

The operation of the 2DEG hydrogen sensor will be described in detail as follows. First, when H_2 comes into contact with Pd, they combine to form PdH_x. The generated PdH_x's potential is lower than the existing Pd. Previous studies used the degree of potential decline given as follows [9]:

$$\triangle \varphi_s = e(\frac{f \bullet Ned}{\epsilon}) \quad (1)$$

The above equation depends on the surface area of the sensor. The Pd deposited through the E-Beam evaporator has a large surface area because it is deposited in the form of nanoparticles. Accordingly, the efficiency in the case of depositing Pd using this process is high. When Pd and H_2 combines and forms PdH_x, the work

function decreases and the bending of the interface increases, thus the electrons are attracted to the 2DEG layer.

The improved conductivity due to the quantitative increase of electrons will reduce the resistance of the sensor. The 2DEG with reduced resistance increases the current flowing between the sensors, and the concentration of hydrogen can be measured through this principle.

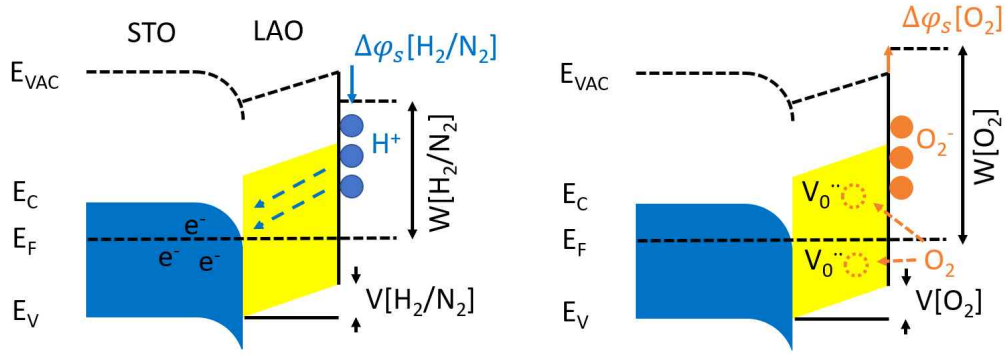

Figure 3. Change of 2DEG potential due to hydrogen bonding When Pd and H_2 combines and forms PdH_x, the work function decreases and results in changing the resistance of the sensor. [9]

Ⅱ.4. Performance of the sensor

There are three main factors that are used to evaluate the performance of the sensor, which are response time, recovery time and sensitivity.

Response time is the time taken to reach a certain percentage of the final value. There are two types of response time: T90 and τ. T90 is the time taken to reach 90% of the final value. τ is the time it takes to reach 63.2% of the final value. Short response time means that the sensor recognizes the change quickly, which means that the sensor's performance is good.

Recovery time refers to the time taken for reuse. It usually means the time it takes to reach 10% of its value after reaching its final value. Sensitivity is defined as follows.

$$\frac{final\ value - initial\ value}{initial\ value} \cdot 100(\%) \quad (2)$$

The larger the sensitivity is, the more sensitive it is to small changes. Although there is no statistical data for expected response time, recovery time, and sensitivity, there is a previous study showing that the performance of the thin-film hydrogen sensor ranged from 2 to 450 s, 20 to 100 s, and 30 to 50% [3]. In each case, it is a value measured at a high-temperature environment of 90°C or higher and a concentration of 1000 pm.

Ⅲ. Experiment

The general aim of the research is divided into two parts. First, it is to measure the performance of the manufactured hydrogen sensor before and after bending. The second aim is to compare the newly manufactured sensor with the sensors that are currently being used. If the manufactured sensor in this research has a good performance, the flexible sensor could be useful in different areas. The following passage explains the methodology of the experiment.

Ⅲ.1. Sensor manufacturing

First, the substrate was fabricated using ALD to form the 2DEG structure. In previous research, it was known that a 2DEG is formed by depositing ALD on a STO layer. In this study, TMA was deposited on the STO through ALD as an aluminum precursor. The process of aluminum oxide deposition in the ALD process is as follows. The entire ALD was conducted under 220°C.

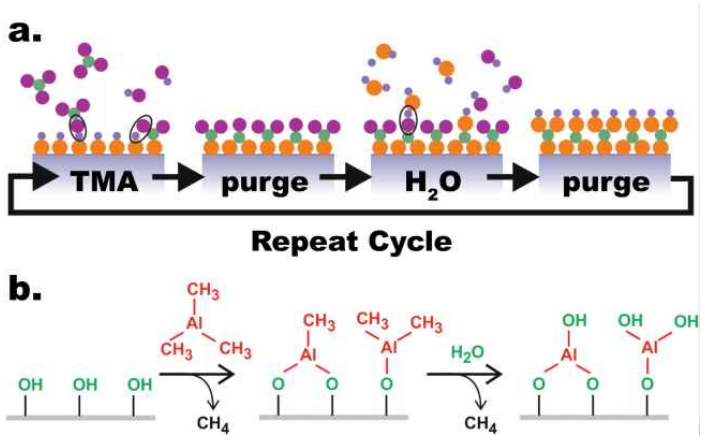

Figure 4. Aluminum deposition in ALD [10]

First, TMA precursors was placed on the STO substrate. Then, methane was separated from the TMA and the precursor was bound to the OH group of the STO substrate. After removing the remaining materials through purge, water was added, which reacted with the left atoms. Then methane escaped and aluminum oxide was attached to the initial STO substrate. In general, a commercially available STO substrate is used in the case of STO/Al_2O_3 2DEG formation. However, in this study, the substrate was manufactured by depositing STO on the Polyimide plate instead of the unstretched STO substrate in order to fabricate a flexible hydrogen sensor respectively.

Figure 5. ALD process equipment

After ALD deposition, Pd was deposited on the E-Beam evaporator. Through this process, it was possible to make a bond between Pd and hydrogen when measuring the concentration of hydrogen. The PdH complex, which was generated by the presence of hydrogen bonds, could induce the work function change in the 2DEG of the sensor and measure the hydrogen concentration.

Figure 6. E-Beam Evaporator used in the study

Ⅲ.2. Hydrogen concentration measurement

In order to understand the hydrogen sensing performance of the completed Polyimide plate hydrogen sensor, the change of the current value according to the hydrogen exposure of the sensor without bending was measured. The response time and the recovery time were measured by changing the current when the sensor was exposed to 5 ppm hydrogen gas and when the hydrogen supply was turned off. The temperature condition was carried out at room temperature, and the experiment was carried out using MMVC3S of MSTECH.

The fabricated sensor was repeated 500 times with a constant radius of curvature. An automatic device was mounted on the step motor to give uniform physical stress. The sensor was then re-deployed and run at 5 ppm hydrogen.

Figure 7. The bending machine used to bend the hydrogen sensor

Figure 8. Measurement of hydrogen concentration using MMVC3S from MSTECH

Ⅳ. Results and Discussion

The data from the manufactured sensor was obtained after the sensor was stabilized. The sensors were stabilized by repeating a process where the sensors were exposed to hydrogen and then stabilized. The following figure shows current-time data of a sensor.

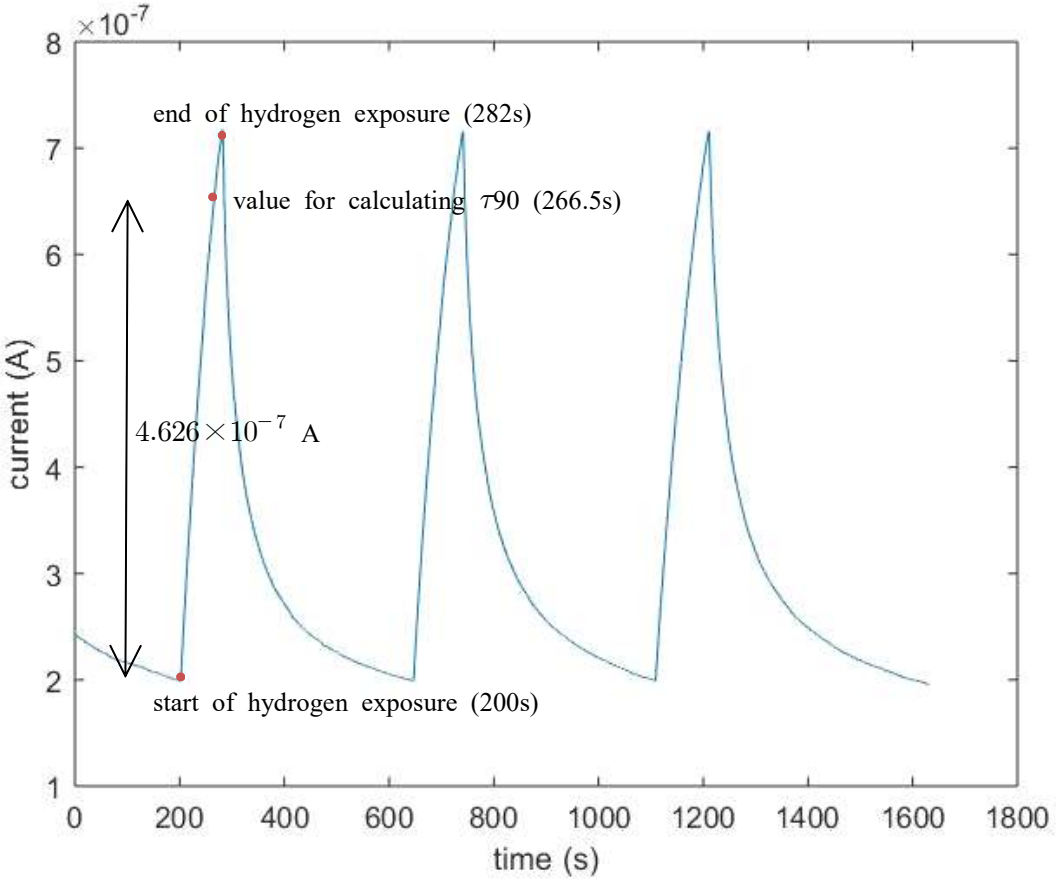

Figure 9. The hydrogen sensor's current graph before bending

The response time, recovery time, and the sensitivity was calculated based on the hydrogen sensor's current data. The next paragraph will explain as an example of calculating

each values.

To calculate the response time(τ90), the following process was done. First, the difference between the current values when the hydrogen exposure started and ended was calculated. From the graph above, the difference of the first peak is 5.14×10^{-7}. Then, 90% of the value was earned, which is 4.626×10^{-7}. Then the time when the current is 4.626×10^{-7} higher than the current at 200 seconds was found, which is 266.5. Then the τ90 was earned, which gave the final value of 66.5 seconds. Like this process, the other values, which are the response time, recovery time and sensitivity, were calculated on the different peaks and after the bending of the sensor was conducted. The following table shows the calculated values of each sensor:

Table 1. The calculated values of the hydrogen sensor before and after bending

Peak Number	Calculated Value	Before Bending	After Bending
1	τ90 (s)	66.5	67
	T90 (s)	150	152
	Sensitivity (%)	285	258
2	τ90 (s)	80	96.5
	T90 (s)	170	160
	Sensitivity (%)	259	250
3	τ90 (s)	76.5	84
	T90 (s)	184	186
	Sensitivity (%)	260	255
Average	τ90 (s)	74.3	82.5
	T90 (s)	168	166
	Sensitivity (%)	259	254.3

In the table, the response time and recovery time were each marked τ90 and T90. The response time and recovery time both showed similar values. The average response time increased from 74.2 seconds to 82.5 seconds, which is 8.17 seconds. The recovery time was

about 167 seconds which did not show major differences before and after the bending process. The Sensitivity were 259% and 254.3% each, which are very close values.

Table 2. The hydrogen sensor's current graph before bending in previous studies [3]

T (°C)	90	110	130	150
90% response time (s)	450	40	7	2
90% recovery time (s)	100	52	20	20
$S_{1000ppm}$ (R_{air}/R_{gas})	50	45	30	50

The graph above shows the response time, recovery time, and sensitivity of a hydrogen sensor developed recently [3]. Compared to the hydrogen sensor in the research above, the recovery time and response time were generally similar, but showed slower time values. However, the measurement of the current values of the sensors from the research above were conducted at 90 ° C, which was not at room temperature. When the manufactured sensor in this paper measured its response time, recovery time and sensitivity at 90 ° C, the response time was 4 times better when the recovery time was 1.6 times longer, and the sensitivity was 5 times better compared to the previous research. Considering these facts, the hydrogen sensor fabricated in this study showed excellent performance.

Ⅴ. Conclusion

In this paper, the main aim was to manufacture a flexible hydrogen sensor that has a performance similar or better than the existing sensors. The flexible hydrogen sensor was manufactured by ALD and E-Beam Evaporator, which made a 2DEG. This 2DEG made it possible for electrons to pass by, which is the key to measuring hydrogen concentration. In this paper, the flexible hydrogen sensor had a response time of 74.3s, recovery time of 168s, and 259% sensitivity before bending. After bending, it had a response time of 82.5s, recovery time of 166s, and 254.3% sensitivity. The statistics provides evidence that are comparable to existing research, because the hydrogen sensor in this paper showed a better statistic in response time, recovery time and sensitivity over previous hydrogen sensors.

As a result, the hydrogen sensor based on the 2DEG fabricated on the Polyimide plate exhibited bending properties and exhibited excellent physical availability almost equal to the flexibility of the Polyimide plate due to the fact that a very thin film was covered on the Polyimide plate. In addition, the superiority of the sensitivity, response time, and recovery time measured by hydrogen exposure after 500 continuous physical stresses did not show a significant decrease. In other words, the performance of the manufactured flexible hydrogen sensor was maintained without deteriorating. In addition, the sensitivity was more than 250%. This is significant because the experiment was conducted under a low hydrogen gas concentration at 5ppm and the measurement was performed at room temperature. Therefore, the manufactured flexible hydrogen sensor showed better performance comapred to the existing sensors. The study implies that the 2DEG-based flexible hydrogen sensor is not only flexible, but also shows great performance, which makes it possible to popularize. So, if this sensor is attached to various surfaces in industries using hydrogen energy such as hydrogen automobile and hydrogen fired battery, it will contribute effectively to prevention of various

accidents by reacting even a small amount of hydrogen gas leakage of 5 ppm level.

However, the limitations of the flexible 2DEG hydrogen sensor have also been partially found in this experiment. One of them is that it is difficult to mass-produce sensors with the same performance. Generally, when comparing the economical feasibility, it is true that the 2DEG manufactured using the ALD method is more affordable than the previous method, but the ALD method has a limited area of the 2DEG interface that can be manufactured at one time. In particular, the initial base current value without exposure to hydrogen is not consistent due to a slight change in the physical properties of the 2DEG when it is produced using other ALD equipment. Also, the base current value of the sensor needed to be checked through the hydrogen removal process after several initial hydrogen exposures. These parts are expected to increase the efficiency of mass production by massing the 2DEG area produced at one time and standardizing the initial base current confirmation work.

It would be another task to produce the flexible hydrogen sensor. When manufacturing the flexible hydrogen sensor by mass production, the initial base current value must be constant. In order for the hydrogen sensors to maintain the initial value, the same ALD process must be performed. This can be controlled by standardizing the ALD process equipment. Also, if the flexible hydrogen sensor is made on a plate that has a big area, it would be easier to keep the characteristic of the sensor : response time, recovery time and sensitivity. Controlling the ALD process equipment and making the sensor on a big planed should be studied further. If these problems are solved, the flexible hydrogen sensor could be used in different situations where the leakage of hydrogen must be checked. The flexible hydrogen sensor has two benefits. First, it can reduce the differences in performance when an outer force is applied. Second, the sensor can be attached in many different places, such as a curved pipe. If the mass production of hydrogen sensors becomes possible, it is expected to decrease accidents related to hydrogen leakage.

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감 사 의 글

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연 구 활 동

- 없음