

Application of Graphene Towards Hydrogen-Gas Sensor

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[Summary]

Since graphene is exposed to the external environment, it is sensitive to surrounding conditions, such as ambient gases. Therefore, it can be developed to high-sensitivity molecular sensors by decorating with receptors that capture target molecules. This article introduces application of graphene to hydrogen-gas sensing devices which have been drawing attention from various fields including energy and medicine. By fabricating graphene into nano-ribbons/meshes and decorating them with palladium as a hydrogen receptor, we successfully developed a high-sensitivity hydrogen-gas sensor capable of detecting hydrogen concentrations below 1 ppm in vacuum. The response of the device electrical resistance to hydrogen gas was different from the results of previous studies, suggesting a possibility of unknown physical phenomenon. Future work to clarify the sensing mechanism and to develop practical device structure and test systems are expected to lead to small, low-power hydrogen-gas sensors.

1 Introduction

Two-dimensional electron systems used for semiconductor devices are inherently located hundreds of nanometers deep from the surface because they are created at the interface of different materials. In contrast, graphene exists on the substrate surface and is exposed to the outside world. Therefore, graphene is sensitive to the external environment, and its physical properties can easily change by altering the surrounding environment, such as ambient gases. Conversely, these changes in graphene's physical properties can be used to evaluate the surrounding environment, which is why graphene is considered to be a promising sensor¹⁾. However, target selectivity is difficult to achieve by using graphene as is, and the sensitivity is not necessarily adequate. Selectively detecting molecules rather than physical quantities such as magnetic or electric fields can be achieved by covering graphene with membranes that only allow passage of the target molecules, or by decorating graphene with receptor substances that capture target molecules. For example, high-sensitivity odor sensors have been proposed by decorating graphene with peptides that have amino acid sequences added to interact with odor molecules²⁾. There are also reports of making biosensors for COVID-19 that are more accurate than antigen tests and faster than PCR tests by decorating graphene with antibodies to the SARS-CoV-2 spike protein³⁾. In principle, it is possible to use graphene in sensors targeting various molecules by changing the receptors.

On the other hand, hydrogen gas is attracting attention as a next-generation energy source for achieving a carbon-neutral society, as well as having applications in medical and disease diagnosis settings. However, hydrogen gas is explosive in air at a wide range of concentrations from 4% to 75%, and also degrades steel materials (hydrogen embrittlement) due to its high material permeability. Being colorless, tasteless and odorless, it is undetectable by human senses. Therefore, detection of hydrogen leaks and measurement of hydrogen concentrations are important at each stage of hydrogen generation, storage, transport, and use. Conventionally, sensors using mass spectrometry are used to measure concentrations accurately, but the equipment becomes large. Small sensors primarily for leak detection either detect changes in the electrical resistance of metal oxide semiconductors due to redox reactions with hydrogen, or detect changes in the electrical resistance of platinum (Pt) wires due to hydrogen gas combustion, or detect changes in the thermal conductivity of gases. However, all operate at high temperatures of several hundred degrees Celsius, which not only increases power consumption but also raises issues about hydrogen explosions, depending on the gas concentration⁴⁾. Therefore, development of hydrogen-gas sensors that are not only fast and sensitive, but are also small, low power and can operate at room temperature is anticipated.

This article introduces the possible applications of graphene, which is nano-fabricated with our unique hydrogen

plasma etching technique, towards hydrogen-gas sensors. Section 1 outlined the potential applications of graphene for molecular sensors and the current state of hydrogen-gas sensing. Section 2 introduces previous research on hydrogen-gas sensing using graphene, and Section 3 discusses our unique methods of preparing samples and evaluating sensor characteristics, as well as experimental results.

2 Hydrogen Gas Sensing Using Palladium-Decorated Graphene

While platinum (Pt) and palladium (Pd) are both known hydrogen receptors, this study only used Pd, which is the more common metal that stores hydrogen even at room temperature. When Pd is exposed to hydrogen gas, hydrogen molecules (H_2) adsorbed on the Pd surface decompose into hydrogen atoms (H), which penetrate into the gaps of the Pd crystals to expand the crystals, forming hydrogenated palladium (PdH_x), and changing the Pd physical properties, such as volume and electrical resistance. The palladium-decorated graphene used in this study detects changes in the Pd physical properties as changes in the graphene electrical resistance. While the presence or concentration of hydrogen gas can be obtained from measuring the physical properties of Pd itself, using Pd-decorated graphene allows for detection of even more subtle changes in Pd properties, meaning even smaller amounts of hydrogen gas can be detected. In addition, it keeps the amount of precious Pd metal low. Furthermore, being able to operate at room temperature without heating for sensing is another major advantage.

It is reported⁵⁾⁻¹¹⁾ that, when Pd-decorated graphene is exposed to mixed gases containing hydrogen at room temperature under atmospheric pressure, the electrical resistance of the graphene device increases according to the concentration of hydrogen gas, and when the hydrogen gas is removed, the electrical resistance returns to its original value. This resistance change is explained by the differences in work functions between graphene, Pd, and PdH_x . In other words, electrons are transferred from graphene to Pd because graphene's work function ($\phi_G = 4.5$ eV) is smaller than Pd's ($\phi_{Pd} = 5.6$ eV) and, therefore, holes are doped into graphene. However, when Pd is hydrogenated, the work function of PdH_x ($\phi_{PdH} = 3.2$ eV) is smaller than the work function of graphene, so the charge flow is reversed, with electrons transferring from PdH_x to

graphene, and, therefore, electrons are doped into graphene. As a result, the electrical resistance of the originally hole-doped graphene devices decreases with Pd adsorption, and increases with exposure to hydrogen.

On the other hand, the sensor functional improvements are expected by nano-structuring of graphene and the processed graphene edges may have functionality in themselves¹²⁾. The length of edges per surface area increases in nanostructures, which improves the sensitivity of detection using edges^{9), 13)}. While devices with graphene nanoribbons (GNRs) processed into nanoscale ribbon shapes arranged in parallel, have lower sensitivity compared to unprocessed cases because the graphene area is smaller and the Pd amount is reduced, there are reports⁸⁾ of faster reaction when hydrogen gas is introduced and recovery when hydrogen gas is removed as well as higher sensor reproducibility.

3 Nanoscale Graphene Hydrogen-Gas Sensor

3.1 Fabricating Nanoscale Graphene

A key feature of this study is nanoscale fabrication of graphene using hydrogen plasma^{14), 15)}. Etching graphene with hydrogen plasma creates hexagonal holes (nano-pits) centered on defects and it is known that the edges are atomically aligned in a zigzag structure^{16), 17)}. With common processing methods, such as oxygen plasma etching or mechanical cutting, the graphene edges are random structures at the atomic scale, and in most cases, the hexagonal rings of carbon atoms are broken into pentagonal or heptagonal rings, or become amorphous^{14), 15)}. In contrast, edges obtained by hydrogen plasma etching have an ideal zigzag shape that maintains the honeycomb structure of graphene¹⁶⁾⁻¹⁸⁾. In nanoscale devices where edge-to-area ratio is larger, it is important to create controlled edges, which is possible using this method with good reproducibility. Additionally, graphene's zigzag edges have electronic states at the energy of the charge neutrality point (Dirac point) due to the breaking of crystal symmetry^{14), 19)}. This is called a zigzag edge state. Therefore, there are hopes that new functionality can be acquired through zigzag edge states while maintaining the graphene properties¹³⁾. This study prepared zigzag graphene nanoribbons (zG NR), which are nanoscale ribbon-shaped graphene terminated with zigzag edges, and zigzag graphene nanomesh (zGNM), which is processed into a mesh by arranging hexagonal nanopits in a

triangular lattice. The zGNM can be interpreted either as a network structure of zGNRs or as an antidot lattice, depending on the perspective or the physical properties being considered. Figure 1(a) shows an atomic force microscope (AFM) image of a zGNR device, and (b) shows an AFM image of a zGNM device. zGNM is believed to have higher industrial value because it is structurally more robust than zGNR as it is connected in a network pattern. Devices used in this study had a total width of 2 to 5 μm , a length of 2 to 3 μm , and a ribbon width (W) of 100 to 200 nm.

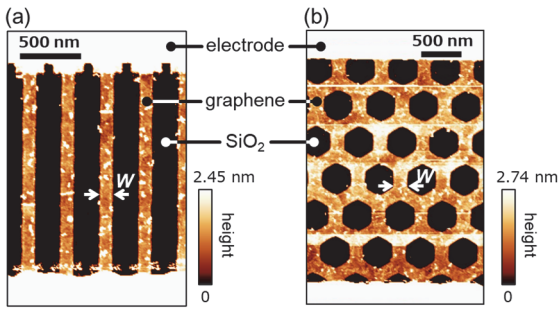


Figure 1 AFM Images of (a) zGNR (approx. 2-nm thick, $W \sim 120$ nm) and (b) zGNM (approx. 2.5-nm thick, $W \sim 200$ nm)

3.2 Pd Decoration of Graphene

Several methods are known for decorating graphene with Pd, and the most common is the thermal evaporation in a vacuum, with which Pd is adsorbed on to the graphene as nanoparticles⁵⁾. When the deposited amount is small, the Pd nanoparticles are isolated. As the deposited amount increases, the Pd nanoparticles begin to cover the entire surface of the graphene, and eventually the Pd begins to exhibit electrical conductivity as a film as the bonds between nanoparticles strengthen. In this study, Pd was vacuum-deposited on zGNR and zGNM by heating Pd with an electric current. Figures 2(a)-(c) show AFM images of the Pd nanoparticle layer for several deposition film thicknesses. Here, the Pd-film thickness was measured using a film thickness meter with a quartz micro-balance placed alongside the graphene sample. Since this corresponds to the film thickness when Pd is attached uniformly to the surface, we express it as the ‘nominal’ film thickness (d_{nom}). In reality, as shown in Figures 2(a)-(c), Pd is deposited as nanoparticles with diameters of 10 to 20 nm, so the surface has irregularities, and the film thickness actually measured by AFM is thicker than d_{nom} . It is known that actual film thickness depends on the Pd deposition rate, too. In this study, Pd was deposited with the deposition rate of approx. 0.9 $\text{\AA}/\text{s}$ with respect to d_{nom} .

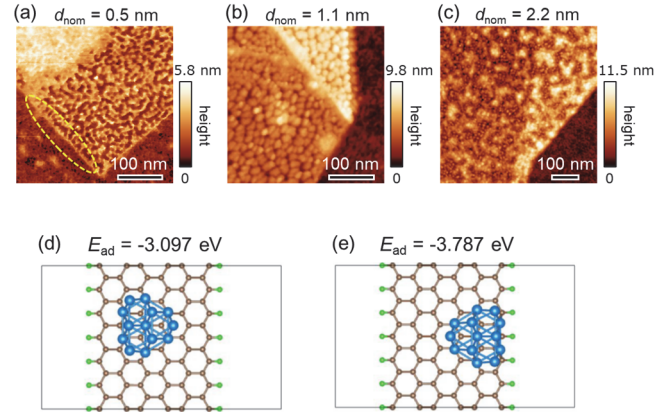


Figure 2 (a)-(c) AFM images of Pd deposition films with nominal thicknesses $d_{nom} =$ (a) 0.5 nm, (b) 1.1 nm, (c) 2.2 nm. As shown by the yellow dotted line in (a), Pd is adsorbed selectively at the edges of graphene flakes. (d)(e) Adsorption structures of Pd clusters (Pd_{13}) placed at center (d) and edge (e) of zGNR, along with their respective adsorption energies (E_{ad}).

As shown by the dotted line in Figure 2(a), the Pd adsorption state at the graphene-flake edges differs from that on the graphene surface. Pd is adsorbed on the surface as nanoparticles, but Pd clumps at edges. This occurs regardless of whether the edge structure is zigzag or armchair-type¹⁴⁾. This selective adsorption of Pd at the edges has also been confirmed by first-principle calculations using the *Fugaku* supercomputer (Figures 2(d)(e)). When calculating the adsorption energy (E_{ad}) of a cluster of 13 Pd atoms (Pd_{13}) adsorbed on zGNR with a width of 0.99 nm, E_{ad} was -3.07 eV on the graphene surface (Figure 2(d)), while -3.78 eV on the zigzag edge (Figure 2(e)). This indicates that Pd clusters are energetically more stable when adsorbed at the edge, meaning they are more likely to adsorb at edges. Since the physical properties of Pd are being measured in hydrogen sensors, the more Pd, the higher the sensor sensitivity in principle. Therefore, the presence of graphene zigzag edges may result in improved sensor sensitivity.

When considering hydrogen sensor applications, it is thought that Pd nanoparticle is better to increase the surface area of Pd exposed to hydrogen gas. Conversely, the Pd film thickness should be as thin as possible to transmit the changes in Pd’s physical properties to graphene more quickly and efficiently. Therefore, in this study, Pd nanoparticles were controlled to have a nominal thickness (d_{nom}) of approx. 0.7 nm to cover the entire surface of the graphene with a single layer.

3.3 The Way to Evaluate Hydrogen-Gas Sensor Characteristics

Figure 3(a) shows the configuration of a hydrogen-gas sensor using Pd-decorated graphene, and Figures 3(b)(c) show the schematic illustrations of the system for evaluating its features. Electrodes are attached to graphene processed into zGNM or zGNR before decoration with Pd. The substrate holding the graphene is highly doped conductive silicon (Si), covered with an insulating surface of SiO₂. The graphene electronic state can be changed by the field effect by applying a gate voltage (V_g) to the Si (Figure 3(a)). This study mainly used a vacuum measurement system (Figure 3(b)). In this setup, only hydrogen gas is introduced into the measurement chamber, which is at a base pressure of ≈ 0.01 Pa, and the introduced amount of hydrogen gas is controlled by the partial pressure (P_{H_2}) of hydrogen gas. On the other hand, most previous studies measured the electrical resistance of the device while flowing hydrogen gas diluted with inert gases such as dry air, nitrogen, or argon into the measurement chamber (Figure 3(c)). In other words, the measurement chamber is at atmospheric pressure. This latter method is closer to actual conditions for detecting leaks to atmosphere, but, since non-hydrogen gas molecules are also present, it is unsuitable for examining the pure reaction between the device and hydrogen.

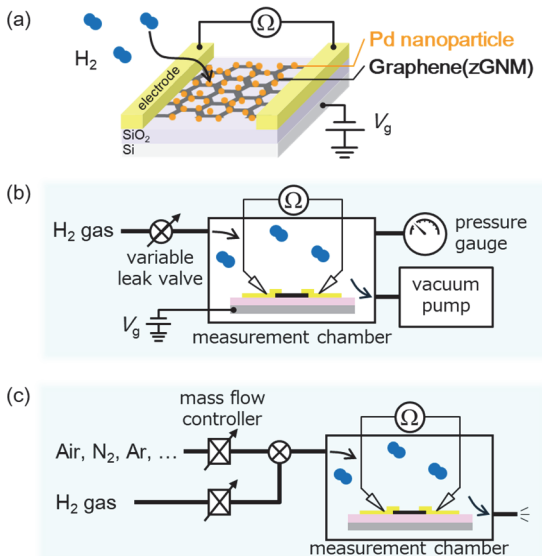


Figure 3 (a) Schematic diagram of Pd-decorated graphene hydrogen gas sensor. (b)(c) Schematic diagrams of hydrogen sensor characterization systems mainly used in this study. Configuration for measurement in vacuum (b), and configuration for measurement at atmospheric pressure (c) used widely in previous research.

3.4 Hydrogen Gas Sensing with Pd-Decorated zGNR/zGNM

Figure 4(a) shows the response by Pd-decorated zGNR (device A) to hydrogen gas. This was measured using the system shown in Figure 3(b). As shown, the electrical resistance (R) of graphene decreases with the introduction of hydrogen gas, and the magnitude of this change depends on the partial pressure (P_{H_2}) of the hydrogen gas. Figure 4(b) shows the ratio of change in R relative to the initial electrical resistance of the device (R_0) before introducing hydrogen gas ($\Delta R/R_0 = (R - R_0)/R_0$) versus P_{H_2} . Although there is some variation, the ratio of change increases as the amount of hydrogen gas increases. This dependency suggests that device A can measure down to $P_{H_2} \leq 0.1$ Pa, corresponding to a concentration of 1 ppm — a sensitivity capable of detecting hydrogen gas in human breath²⁰). Consequently, we found that a hydrogen-gas sensor with high sensitivity even at room temperature can be realized using our fabrication technique.

In the case of device A (Figure 4(a)), R continued changing even at 2 minutes after hydrogen gas was introduced, and had not returned to the initial value even at 5 minutes after stopping introduction of hydrogen gas. However, since R starts changing within 0.5 s after introducing hydrogen gas into the measurement chamber, the continuing sensor response is not because the chemical reaction rate is slow. The continuing change in R is possibly because the amount of hydrogen gas contributing to the reaction per unit time is small, requiring time for the entire device to change. Decreasing the device size or improving its structure might lead to a sensor where R saturates within a practical measurement time. In fact, a device, R of which saturates within 50 s after introducing hydrogen gas, had also been obtained.

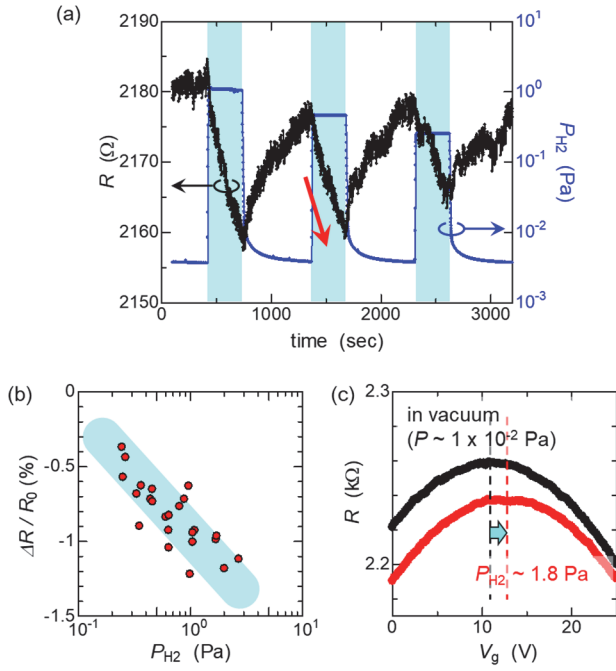


Figure 4 (a) Changes in R of Pd-decorated zGNR (device A) when hydrogen gas is introduced into measurement chamber in vacuum. The electrical resistance of device A (black) is on the left axis, and P_{H_2} is on the right axis. (b) $\Delta R/R_0$ of device A versus P_{H_2} . (c) V_g dependency of R for device A. The black line shows measurement in vacuum ($P \sim 1 \times 10^{-2}$ Pa), and the red line shows measurement at $P_{H_2} \sim 1.8$ Pa.

Figure 4(c) shows the measurement of the V_g dependency of R for the same device A to investigate the origin of these electrical resistance changes. Graphene has linear energy dispersion with the charge neutrality point (Dirac point) as its intersection. Therefore, the V_g dependency of R reaches its maximum at $V_g = V_{\text{Dirac}}$ corresponding to the Dirac point energy. Figure 4(c) shows that as hydrogen gas is introduced, not only does R decrease, but V_{Dirac} also shifts in the positive direction. This behavior was reproducible with the introduction and removal of hydrogen gas, and was similarly observed in all measured samples regardless of the graphene shape. The fact that V_{Dirac} is positive even before hydrogen-gas introduction indicates that device A was originally hole-doped, and the increase in V_{Dirac} in the positive direction at hydrogen-gas introduction suggests that hydrogen gas causes hole-doping in graphene. In other words, this study found that the Pd-decorated graphene used in this study is hole-doped by hydrogen-gas introduction, resulting in decreased electrical resistance at $V_g = 0$.

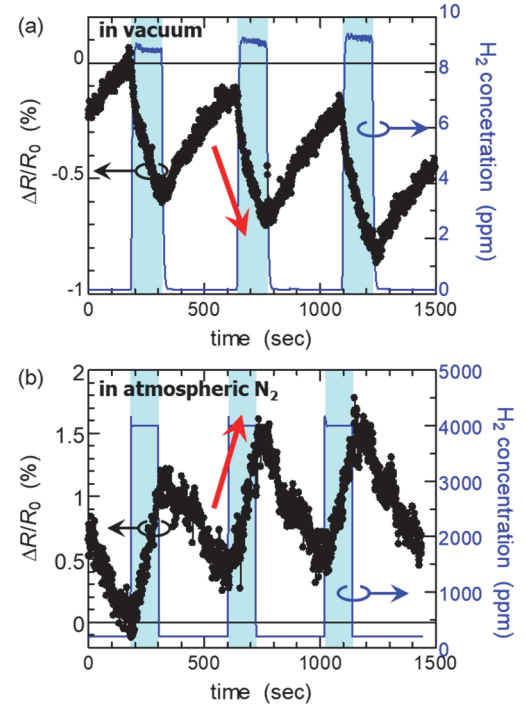


Figure 5 Comparison of hydrogen-gas response of Pd-decorated zGNR (device B). (a) Measurement in vacuum, (b) Measurement in nitrogen atmosphere at atmospheric pressure.

As a result, the Pd-decorated graphene devices fabricated in this study can be expected as highly sensitive hydrogen-gas sensors operating at room temperature. However, the result where R decreases due to hydrogen gas is opposite to that reported in previous research. Differences between this and previous studies lie in the graphene processing method and the measurement environment explained in section 3.3. While this study made measurements in vacuum, previous studies measured at atmospheric pressure. Therefore, we compared the reaction for the same device in vacuum and using hydrogen gas diluted with dry nitrogen gas at atmospheric pressure. Figures 5(a) and (b) show the results for Pd-decorated zGNR (device B) in vacuum and at atmospheric pressure, respectively. These figures show that, similar to Figure 4(a), R decreases with the introduction of hydrogen gas in vacuum, but increases in nitrogen gas at atmospheric pressure, which is consistent with previous research. Additionally, the hydrogen-gas concentration needed to obtain approximately 0.5% of $\Delta R/R_0$ was about 9 ppm (0.9 Pa) in vacuum, but approximately 2000 ppm (0.2%) at atmospheric pressure. These results show a decreased sensitivity of more than 2 orders of magnitude. Since the dependency of V_g on R was not measured at atmospheric pressure, it is impossible to clearly demonstrate the doping effect shown in Figure 4(c). However, since we

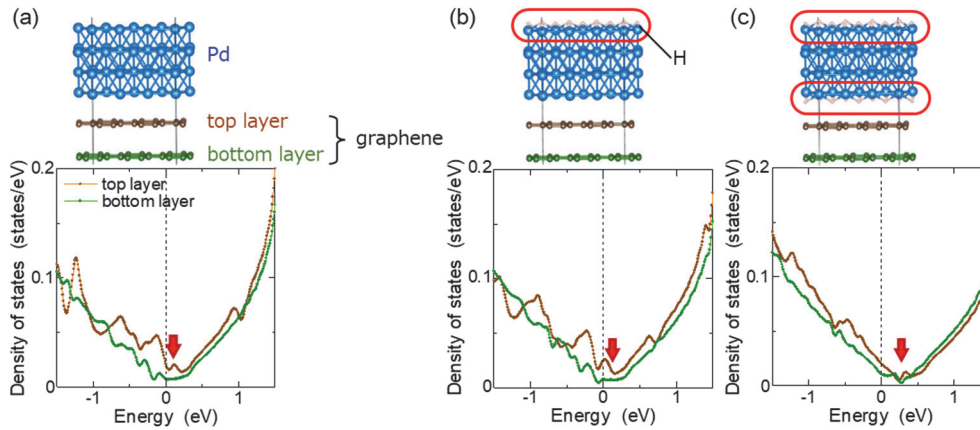


Figure 6 Cross-sections of the system where Pd clusters adsorbed on bilayer graphene (top panel) and electron density of states of graphene (bottom panel). (a) Without hydrogen atoms, (b) Hydrogen atoms adsorbed on top surface of Pd cluster, (c) Hydrogen atoms adsorbed on both top and bottom of Pd cluster.

confirmed that the device is hole-doped even at atmospheric pressure, the increase in R caused by hydrogen-containing gas is likely because electrons are doped into the graphene.

Consequently, we think the different results between our study and previous studies is not due to the device itself, but is rather due to differences in the measurement environment, specifically whether measurement is in vacuum or at atmospheric pressure. Generally, a vacuum environment increases adhesion between graphene and substrate, or removes impurities adsorbed between graphene and substrate or on the graphene surface. However, it is difficult to imagine that such effects would change the chemical reaction. In this case, the significant effect can be considered as that only hydrogen gas is introduced at in-vacuum measurement, whereas gas molecules other than hydrogen are also present at atmospheric pressure measurements. Very recent measurements introducing hydrogen gas diluted with dry air in vacuum also resulted in increased electrical resistance of the device. Considering these factors, it appears that the presence of gas molecules other than hydrogen is related significantly to the behavior of the device resistance. When hydrogen gas comes into contact with Pd-decorated graphene, holes are doped into the graphene and the electrical resistance of the device decreases, but this reaction is reversed when other gas molecules are involved. Although previous studies have used dry air, nitrogen, and argon to dilute hydrogen, the direction of the reaction is the same in all cases. Therefore, the identity and role of gas molecules other than hydrogen that contribute to increased electrical resistance of the device remain unknown and are a subject for future research.

To verify the possibility of hole doping by hydrogen molecules in Pd-decorated graphene, first-principle calculations were performed using *Fugaku* to investigate the charge transfer to graphene when hydrogen molecules that decomposed into atoms on the Pd surface are adsorbed onto Pd. *Fugaku* simulated a system where Pd atoms are adsorbed in layers on bilayer graphene. The electron density of states of graphene was calculated when hydrogen atoms are adsorbed on the layered Pd cluster surface, while simultaneously calculating the charge transfer from hydrogen-adsorbed Pd to graphene using the Bader method. Figure 6 shows the calculated model (top panel) and the corresponding electron density of states (bottom panel). Since the Dirac point is the intersection of linear energy dispersion, the energy at which the electron density of states is minimized corresponds to the Dirac point (arrow in Figure 6). Figure 6(a) shows the case without hydrogen, Figure 6(b) shows the case when hydrogen atoms are adsorbed on the top surface of the layered Pd cluster, and Figure 6(c) shows the case when hydrogen atoms are adsorbed on both the top and bottom surfaces of the layered Pd cluster, the latter of which is in contact with graphene. Although the fine structure of the electron density of states differs in each case, when hydrogen atoms are adsorbed on the top surface of the Pd cluster, the energy of the Dirac point does not change much compared to the case without hydrogen. However, when hydrogen atoms are intercalated between the Pd cluster and graphene and adsorbed on the bottom surface of the Pd cluster, the results clearly show that the energy of the Dirac point shifts toward the higher energy side. This corresponds to hole-doping in graphene, which is consistent with our experimental results.

Although graphene hole-doping also occurs according to the height of the Pd cluster from the graphene surface, the size of the charge transfer calculated by the Bader method is more than an order of magnitude larger when hydrogen is adsorbed on the bottom surface of the Pd cluster than when there are no hydrogen atoms, even at the same Pd-to-graphene distance. Moreover, an experiment showed there was no change in the device electrical resistance when introducing nitrogen gas without hydrogen. Considering all these observations, the observed hole-doping is likely a reaction specific to hydrogen gas and is possibly an effect of the hydrogenation of the bottom surface of the Pd cluster.

4 Conclusions

This study discussed the potential use of Pd-decorated graphene as a hydrogen-gas sensor using Pd as a hydrogen-gas receptor. By depositing Pd on graphene as zGNR and zGNM nano-fabricated by hydrogen plasma etching technique, we successfully created a highly sensitive hydrogen-gas sensor capable of detecting hydrogen concentrations below 1 ppm even at room temperature. Future work requires reducing variations in the electrical resistance (R) of the sensor device and investigating and improving sensitivity and response-speed limiting factors. On the other hand, in the aspect of basic research, we discovered that the R direction of change differs according to the hydrogen gas introduction environment. This discovery challenges previously established theory about the hydrogen-gas detection mechanism of Pd-decorated graphene based on the work functions of graphene, Pd, and PdH_x. Future research is required to clarify this mechanism.

Although this study targeted development of a hydrogen-gas sensor using Pd as a hydrogen receptor, sensors for other molecules could be created by changing the receptor. This research offers various sensor applications such as biosensors and odor sensors.

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- 20) The concentration of hydrogen gas in human breath is typically around 5 ppm.

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