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Insertion of a Graphene Oxide Layer into a Cu/SiO₂/Pt Structure to Overcome Performance Degradation in a Vaporless Environment

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Abstract: A Cu/SiO₂/Pt structure is usually used to study the resistive memory properties of an electrochemical resistive random access memory. It can be reversibly switched between low- and high-resistance states by using DC voltages in the atmosphere. However, its resistive switching behavior disappears in a vaporless environment because no conducting filaments can be formed within the Cu/SiO₂/Pt structure. This study inserted a graphene oxide (GO) layer to fabricate a Cu/GO/SiO₂/Pt structure that could be resistively switched in a vaporless environment. The X-ray photoelectron spectra depth profile of the Cu/GO/SiO₂/Pt structure showed that oxygen-related groups of the GO film reacted with the Cu electrode. The GO film assisted Cu ionization in a vaporless environment, and Cu ions could migrate in an electrical field to the Pt electrode. Cu conducting filaments were formed and ruptured by different polarity voltages, and the resistance of the Cu/GO/SiO₂/Pt structure could be reversibly switched in a vaporless environment. A schematic model was proposed to explain the switching mechanisms in the atmosphere and a vaporless environment.

Keywords: graphene oxide; RRAM; vaporless environment

1. Introduction

After the development of consumer electronic products, semiconductor non-volatile memory (NVM) has become increasingly prevalent due to its high operation speed, low power consumption, and shock immunity. Flash memory is now mainstream for NVM devices. However, with continuous device scaling, physics limitations cause severe reliability issues. Therefore, many novel devices, such as magnetoresistive random access memory [1], ferroelectric memory [2], phase change memory [3], and resistive random access memory (RRAM) [4], have been proposed as next-generation NVM devices. Among these devices, RRAM, with a sample metal/insulator/metal structure and resistive switching behavior-induced applied voltages, is the most promising candidate.

The resistance of a RRAM device can be reversibly switched between a high-resistance state (HRS) and a low-resistance state (LRS) using applied voltages. The resistive switching behaviors of RRAM devices are influenced by its material group, defect status, and device structure [5,6], and the switching mechanisms are usually dominated by valence change mechanisms [7], thermochemical reactions [8], and electrochemical reactions [9]. The Cu/SiO₂/Pt structure is a typical electrochemical reaction RRAM, and we previously investigated its resistive switching (RS) properties [9]. Cu ions were dissolved from the Cu electrode; thus, the formation and rupture of Cu conducting filaments

were responsible for the reversible RS mechanism. One of the most severe issues is large switching dispersion, which would cause a small switching margin and a complex circuit for memory applications. Therefore, several methods have been proposed to reduce the occurrence of switching dispersion. Cu nanoparticles (Cu-NPs) were embedded into the SiO₂ layer of the Cu/SiO₂/Pt structure to enhance the local electrical field near Cu-NPs during the forming process, which reduced the switching dispersion [10]. In our previous work [11], a graphene oxide (GO) layer with a folded, layered structure was used to limit the number of formation sites of Cu conducting filaments, which reduced the switching dispersion in the atmosphere [12]. Although the Cu/SiO₂/Pt structure had good RS properties in the atmosphere, it had poor RS properties in a vaporless environment [13], which could have caused problems after device packaging. We have already conducted a study in which we fabricated a Cu/Cu_xO/SiO₂/Pt structure to investigate its RS in a vaporless environment [13]. The Cu_xO layer in the Cu/Cu_xO/SiO_x/Pt structure helped the dissolution of Cu ions from the Cu electrode into the SiO_x layer, enabling electrochemical RS in a vaporless environment.

Graphene is a two-dimensional material with superior electrical and mechanical properties [14], making it a very attractive material for many applications such as integrated circuits [15] and mechanical resonators [16]. Many methods, such as chemical vapor deposition [17] and reduced GO, attempt to fabricate low-price and large-area graphene layers. Oxygen functional groups within GO layers are influenced by synthesis methods and process parameters [18]. Therefore, GO is suitable for many applications. GO can be a surfactant due to its hydrophobic/hydrophilic property [19]. GO nanosheets are also a highly efficient and universal demulsifier [20]. In this study, a GO layer is inserted to fabricate a Cu/GO/SiO₂/Pt structure. The GO film with numerous oxygen-related groups reacted with the Cu electrode to help the dissolution process of Cu ions into the SiO₂ layer. Therefore, the Cu/GO/SiO₂/Pt structure can be resistively switched in a vaporless environment.

2. Experimental Procedures

A 20 nm SiO₂ film was deposited on a Pt-coated substrate (Pt/Ti/SiO₂/Si) by radio-frequency sputtering at room temperature. Then, a 200 nm thick Cu film was deposited using a thermal evaporator to fabricate the $Cu/SiO_2/Pt$ structure (the control sample). The GO suspension was prepared according to the modified Hummer's method [21]. In brief, the preparation procedures of the GO suspension are described as follows: (1) First, 0.5 g of graphite powder and 3 g of potassium permanganate (KMnO₄) were added into 25 ml of concentrated sulfuric acid (H₂SO₄) and then were stirred for 6 hours while keeping the solution temperature less than 25 °C. (2) Next, 200 ml of deionized (DI) water was added and the mixture was stirred for 30 min. Then, 10 ml of hydrogen peroxide (H_2O_2) was added and the mixture was stirred for 30 min. (3) The solution was filtered by a filter paper and then DI water and hydrochloric acid (HCl) were used to wash the filtered slurry several times. After drying the filtered slurry, the graphite oxide powder was obtained. (4) Subsequently, 60 mg of graphite oxide powder, 10 ml of DI water, and 20 ml of alcohol were mixed and stirred. The resulting solution was exfoliated into the GO using an ultrasonic bath for 1 hour. After that, the solution was centrifuged at 5500 rpm for 30 min and then the upper clear part was taken out as the GO suspension. Oxygen functional groups within the GO were influenced by the synthesis method, process parameters, and precursor materials [22,23]. The GO suspension was filtered through a 0.2 µm syringe filter and dropped onto SiO₂/Pt substrates to deposit a GO layer through spin coating. The transmission electron microscope (TEM) image shows that the 5 nm GO layer was uniform and had a folded, layered structure [11]. A 200 nm Cu layer was then deposited to form the $Cu/GO/SiO_2/Pt$ structure (the GO sample). The device area, patterned by a metal mask, was 5×10^{-5} cm². The electrical measurements were performed on an HP 4155B semiconductor parameter analyzer in the atmosphere and an N_2 ambient. The sweep voltage was applied on the Cu top electrode, whereas the Pt bottom electrode was grounded. The chemical binding of the GO layer was characterized through X-ray photoelectron spectroscopy (XPS, JEOL JAMP-9500F, JEOL, Tokyo, Japan).

3. Results and Discussion

Figure 1 depicts the C1s XPS spectra of the GO layer on the SiO₂/Pt structure. The peaks of sp² (aliphatic, C–C), sp³ (aromatic, C–C), hydroxyl (C–OH)/epoxy (C–O–C), carbonyl (C=O), and carboxyl (O=C–OH) were observed at 284.52, 285.08, 286.39, 287.06, and 288.57 eV, respectively [21]. Therefore, a GO layer with many oxygen-related groups was deposited effectively. Figure 2a depicts the XPS depth profile of the GO sample. The carbon peak indicates that the GO layer was between the Cu and SiO₂ layers. Figure 2b depicts the C1s XPS spectra of the GO layer between the Cu and SiO₂ layers. The proportions of C-containing components calculated from the XPS spectra of Figures 1 and 2b are listed in Table 1. After the Cu electrode deposition, the number of oxygen functional groups within the GO layer decreased. The reduction process of reduced graphene oxide can be achieved through chemical, thermal, or electrochemical reduction [21] because the oxygen functional groups within the GO layer are unstable. Therefore, the oxygen functional groups within the GO layer would react with the Cu electrode due to chemical reduction or thermal reduction during the Cu deposition process. Hence, some oxygen functional groups reacted with the Cu electrode to form the Cu_xO interface.



Figure 1. X-ray photoelectron spectroscopy (XPS) spectra of the graphene oxide (GO) layer on the SiO_2/Pt structure.



Figure 2. (**a**) XPS depth profile of the Cu/GO/SiO₂/Pt structure. (**b**) C1s XPS spectra of the GO layer between the Cu and SiO₂ layers.

Peak	Assignment	GO on SiO ₂	GO between Cu/SiO ₂
C-C	sp ² carbon	19.0%	51.8%
C-C	sp ³ carbon	20.5%	28.5%
C-OH/C-O-C	hydroxyl/epoxide	18.2%	5.1%
C=O	carbonyl	29.6%	5.5%
O=C-OH	carboxyl	12.7%	9.1%

Table 1. The calculated proportion of C-containing components of the C1s spectra.

The RS properties of the Cu/SiO₂/Pt structure in the atmosphere were thoroughly investigated in our previous study [9], and the resistance of the $Cu/SiO_2/Pt$ structure can effectively be reversibly switched between an LRS and an HRS by bipolar applied voltages. However, Figure 3a indicates that the control sample cannot be reversibly switched by applied voltages in the N2 ambient. Initially, the current of the device increased as the applied voltage increased. When the applied voltage was increased to 6 V, the current increased abruptly and the resistance state was switched from the initial-resistance state (IRS) to the LRS, known as the forming process. To prevent permanent damage, a current compliance of 1 mA was used. The applied voltage was then swept in the negative polarity to switch the resistance state to an HRS. However, resistance states cannot be switched back to the HRS. Figure 3b shows that RS properties of the GO sample can be found in the N₂ ambient. The RS was initialized by the forming process, and the resistance state switched from the IRS to the LRS. The resistance state was then switched from the LRS to the HRS using a negative voltage, and this was called the "Reset" process. Subsequently, the resistance state could be switched from the HRS to the LRS by a positive voltage, which was called the "Set" process. The resistance state could be reversibly switched between the HRS and the LRS by the Reset and the Set processes. For the two samples, the forming processes were quite different. Due to the dissolution of Cu ions into the SiO_2 layer of the GO sample, more defects within the SiO₂ layer caused a larger leakage current. Therefore, the IRS resistance of the GO sample was lower than that of the control sample. In addition, the forming voltage of the control sample was approximately 6 V, whereas that of the GO sample was approximately 2.2 V. For the GO sample, Cu ions were dissolved into the SiO₂ layer, and thus only 2.2 V was initially required to form Cu conducting filaments. For the control sample, the abruptly increased current of 6 V was due to the dielectric breakdown and not to the connection between the top and bottom electrodes by Cu conducting paths. Therefore, after the forming process, the resistance state of the control sample could not be switched back to the HRS, which was unsuitable for RRAM applications.



Figure 3. (**a**) Resistive switching (RS) properties of the control and (**b**) GO samples in the N₂ ambient. LRS: low-resistance state; IRS: initial-resistance state; HRS: high-resistance state.

The fitting results from the current–voltage characteristics of the GO sample in the N_2 ambient (not shown) indicate that the conduction mechanisms of the HRS and the LRS were both dominated

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by ohmic conduction. The temperature coefficient of the LRS resistance for the GO sample in the N₂ ambient was 4.0×10^{-3} K⁻¹ (not shown), which is close to the value of the Cu bulk. Therefore, the RS was dominated by the rupture and formation of Cu filaments. In the RS process, Cu ions were dissolved from the Cu electrode into the SiO₂ layer, and then Cu ions migrated to the Pt electrode and were reduced on the Pt electrode. Subsequently, the Cu filaments grew from the Pt electrode toward the Cu electrode; thus, the resistance state was switched from the IRS to the LRS [24]. However, the Cu ionization process must be assisted by H₂O molecules [25]. Therefore, the control sample did not possess the RS property in the N₂ ambient.

Figure 4 shows the cumulative probabilities of switching the parameters of the GO sample in the atmosphere and the N₂ ambient. Figure 4a indicates that the HRS/LRS ratios of the GO sample in the atmosphere and the N_2 ambient were 3176 and 4647, respectively. The HRS exhibited a higher dispersion in the N₂ ambient than in the atmosphere, whereas the LRS demonstrated similar values and dispersion in the two ambients. This is because the LRS resistance was dominated by the current compliance. Figure 4b indicates that the Reset voltages were almost the same in the two ambients, whereas the Set voltage in the N_2 ambient was larger than that in the atmosphere. In addition, the Set voltage exhibited a much larger dispersion in the N_2 ambient than in the atmosphere. Because the LRS resistance demonstrated more stability than the HRS resistance did, the Reset voltage was also more stable than the Set voltage was in the N_2 ambient. All switching parameters in the N_2 ambient exhibited a larger dispersion than those in the atmosphere, especially the HRS resistance and the Set voltage. Although the GO layer can assist the dissolution process of Cu ions into the SiO₂ layer in the N_2 ambient to enable RS, the dissolution process of Cu ions should be facilitated by the additional O_2 and H_2O molecules in the atmosphere [25]. The GO sample would then easily form Cu conducting filaments in the atmosphere. Therefore, the GO sample had a lower Set voltage in the atmosphere than in the N₂ ambient. Fewer possible paths were formed and can be selected for successive RS cycles, which reduced the switching dispersion [12]. Different statuses of oxygen functional groups within the GO layer [26] would cause different statuses of the Cu_xO interface. More oxygen functional groups within the GO layer would lead to a thicker Cu_xO layer. However, a thicker Cu_xO layer might retard the migration of Cu ions and a thinner Cu_xO layer might not assist Cu dissolution effectively. Therefore, an optimized Cu_xO thickness within the GO sample would have a better RS property.



Figure 4. Cumulative probabilities of switching parameters of the GO sample in the atmosphere and the N₂ ambient: (**a**) high-resistance state (HRS) and low-resistance state (LRS) resistances; (**b**) V_{Set} and V_{Reset} .

Figure 5 demonstrates that the GO sample can be reversibly switched in the atmosphere and the N_2 ambient for more than 3000 cycles. The HRS/LRS ratios exceeded three orders of magnitude, indicating that the GO sample exhibited stable switching and a large switching margin. Figure 6 indicates that the retention characteristics of the GO sample in the atmosphere and the N_2 ambient

were more than 10^4 s at room temperature. In addition, the HRS/LRS ratio can maintain more than three orders of magnitude for 10^4 s. The HRS resistances in the two ambients kept almost the same, while the LRS resistance degraded faster in the atmosphere than in the N₂ ambient. Tsuruoka et al. proposed that the migration speed of Cu ions in a SiO₂ layer decreases with a reduction in the ambient H₂O pressure [25]. Therefore, Cu atoms diffuse out more easily in the atmosphere than in the N₂ ambient. Hence, the LRS resistance degraded faster in the atmosphere than in the N₂ ambient. The GO sample had better retention characteristics in the N₂ ambient, making it more suitable for nonvolatile memory application.



Figure 5. Endurance characteristics of the GO sample in (a) the atmosphere and (b) the N₂ ambient.



Figure 6. Retention characteristics of the GO sample in (a) the atmosphere and (b) the N₂ ambient.

Figure 7 proposes the RS model of the GO sample in the atmosphere and the N₂ ambient. A Cu_xO layer is formed by a chemical reaction between the Cu and GO layers. Hence, the Cu_xO layer assists the Cu ionization process, causing the GO sample to be resistively switched in the N₂ ambient. When the GO sample is in the atmosphere, H₂O molecules are absorbed in the GO and SiO₂ layers. The migration speed of Cu ions in a SiO₂ layer increases with the increasing ambient H₂O pressure [25]. The Cu ionization process is assisted by both the Cu_xO layer and moisture. Therefore, the GO sample has a lower switching dispersion in the atmosphere than in the N₂ ambient.



Figure 7. RS model of the GO sample in the atmosphere and the N₂ ambient.

4. Conclusions

A Cu/SiO₂/Pt structure showed the resistive switching property in the atmosphere. Oxygen and H₂O molecules reacted with the Cu metal to form a Cu_xO layer during the forming process, and then the Cu_xO layer assisted the Cu ionization process. Thus, Cu ions migrated through the SiO₂ layers under an electric field. Hence, the formation and rupture of Cu conducting filaments are responsible for the RS. However, the Cu/SiO₂/Pt structure cannot form a Cu_xO layer in a vaporless environment and thus did not show the RS property. A GO layer was inserted into the Cu/SiO₂/Pt structure to fabricate the Cu/GO/SiO₂/Pt structure. The GO layer was full of oxygen-related groups and hence reacted with the Cu electrode to form the Cu_xO layer. The Cu_xO layer assisted the Cu ionization process, and Cu conducting filaments could thus be formed within the SiO₂ layer, reversibly enabling RS in a vaporless environment. The RS of the GO sample in the atmosphere was also characterized. Although the GO sample had slightly poorer RS in the N₂ ambient than in the atmosphere, it had high endurance (> 3000 cycles) and high retention (> 10⁴ s). Therefore, the Cu/GO/SiO₂/Pt structure is suitable for next-generation NVM applications.

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