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Three Dimensional Floating Gate Memory with Multi-layered Nanodot Array Formed by Bio-LBL

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1. Introduction

Nanodot-type floating gate memories have attracted much attention due to their high reliability and low operation voltage. Our group has developed the fabrication of the nanodot-type floating gate memory by utilizing ferritin. We achieved the high-density nanodot array with 8.0×10^{11} cm^{-2} by utilizing ferritin. However, higher adsorption density of nanodots than 10^{12} cm^{-2} is required for future memory devices. To realize high-density nanodot array with over 10^{12} cm^{-2}, we have investigated three dimensional floating gate memory with multi-layered nanodot array formed by Bio-Layer-By-Layer (Bio-LBL).

Figure 1 shows the structure of the floating gate memory with multi-layered bio-nanodot (BND) array. Bio-LBL is the stacking process of nano particle utilizing selective adsorption and biomineralization of Titane-Binding Ferritin (TBF), as shown in Fig.2. [2] First, TBF adsorbs onto Ti, Ag, and Si. After the adsorption, SiO_2 is formed on the surface of TBF by biomineralization. Multi-layered TBF array with different material BND can be formed by repeating selective adsorption and biomineralization.

In this study, we demonstrate the *C-V* characteristic of the floating-gate MOS capacitor with multi-layered BND array formed by Bio-LBL.

2. Experimental

We used p-type Si (100) substrate with 3-nm-thick SiO_2 as a tunnel oxide. TBF accommodating BND was adsorbed onto the SiO_2 by dropping 0.5 mg/ml TBF solution on the SiO_2. An excess TBF solution was rinsed with 50 mM Tris-HCl solution (TBS). Samples were dipped in a mixture solution of 1-mM HCl and tetramethoxysilane for 30 min to form an interface layer of SiO_2 (TMOS-HCl). After the formation of the interface layer, the samples were rinsed with TBS and water and then dried by centrifugation at 4000 rpm for 1 min. After drying, interface layer was treated by ultraviolet irradiation in ozone ambient (UV ozone treatment) at 115 ^oC for 10 min. After UV ozone treatment, TBF was adsorbed onto the interface layer. A multilayered-TBF array was formed by repeating TBF adsorption and SiO_2 formation procedures. After stacking TBF arrays by Bio-LBL, the outer protein of TBF was completely removed by UV ozone treatment at 115 ^oC for 60 min. 20-nm-thick SiO_2 was deposited on tunnel oxide as a control oxide by plasma-enhanced chemical vapor deposition. Al electrodes were formed and the fabricated MOS structures were annealed in a reductive gas (N_2:H_2 = 9:1) at 450 ^oC for 1 h.

3. Results and Discussion

Figure 3 (a), (b), and (d) show the SEM images of TBF adsorbed on SiO_2 layers. High-density TBF arrays were formed on SiO_2 layers. Figure 3 (c) and (e) show the SEM images of SiO_2 interface layer formed on the first TBF array and the second TBF array, respectively. Figure 4 shows a cross section TEM image of the MOS structure with double-layered Fe-BND array. The double-layered Fe-BND array was clearly seen to be embedded in SiO_2 gate oxide.

Figure 5 shows the *C-V* characteristics of the MOS capacitors with multi-layered Fe-BND array. The *C-V* curves with large hysteresis were observed. The hysteresis was caused by electron injection and hole injection into Fe-BND. Figure 6 shows the memory windows of the MOS capacitors with multi-layered Fe-BND array. The memory window increased with increasing stacked Fe-BND arrays. The adsorption density of Fe-BNDs on tunnel oxide was increased by stacking Fe-BND arrays and the numbers of electrons and holes injected into Fe-BNDs increased.

Figure 7 and Figure 8 show the *C-V* characteristic and memory windows of the MOS capacitor with multi-layered Ni-BND, respectively. The *C-V* characteristics with large hysteresis were also observed. In addition, the memory window increased with increasing stacked Ni-BND arrays. The memory window of MOS capacitor with Ni-BND array is larger than that of Fe-BND array, as shown in Fig. 9. The work function of Ni is 5.2 eV and deeper than that of Fe, 4.5 eV, as shown in Fig.10. Therefore, it is difficult for electrons to escape from Ni nanodot and large amount of electrons were confined to BND and large memory window was observed.

We fabricated the floating-gate MOS capacitor with multi-layered nanodot array by utilizing Bio-LBL for the first time. The memory window of the MOS capacitor increased in proportion to the density of TBF adsorbed on tunnel oxide. The technique is promising for the fabrication of high-performance nanodot-type floating gate memory at low temperature.

4. Acknowledgement

Authors would like to thank Dr. K. Shiba in Japanese Foundation for Cancer Research for the help of Bio-LBL method.

[1] A. Miura et al., *J. Appl. Phys*., vol. 103, 074503 (2008)
[2] K. Sano et al., *J. Am. Chem. SOC*., vol. 128, No.5 (2006)

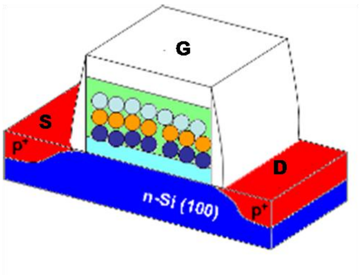

Fig.1. Structure of floating gate memory with multi-layered BND array

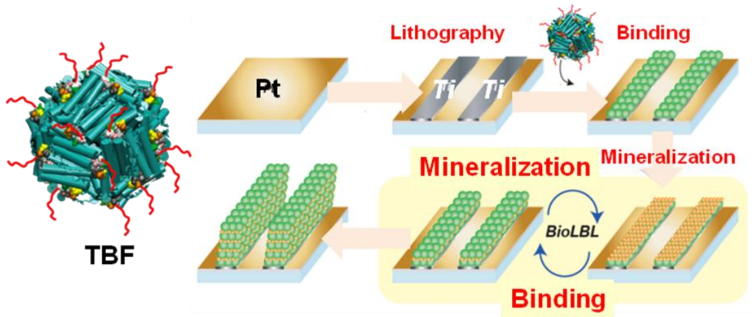

Fig.2. Process flow of Bio-LBL method.

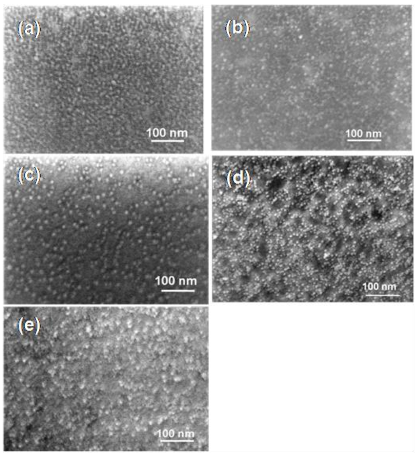

Fig.3. SEM images of (a) TBF adsorbed on tunnel oxide. (b) TBF adsorbed on first SiO_2 interlayer. (c) First SiO_2 interface layer. (d) TBF adsorbed on second SiO_2 interface layer. (e) Second SiO_2 interface layer

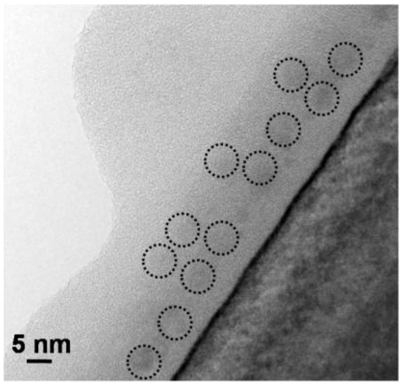

Fig.4. A cross section TEM image of the MOS capacitor with multi-layered

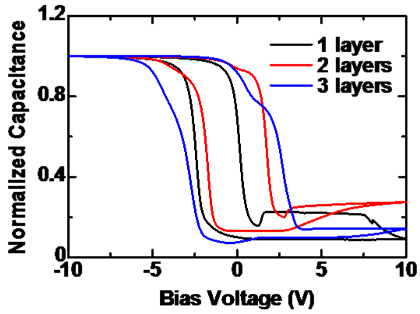

Fig.5. *C-V* characteristics of the MOS capacitors with multi-layered Fe-BND array

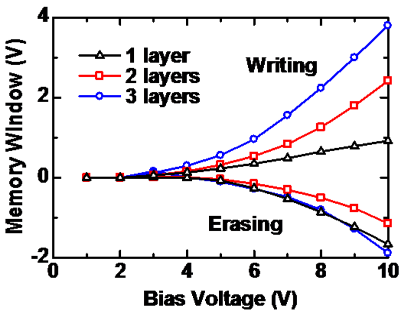

Fig.6. Memory windows of the MOS capacitors with multi-layered Fe-BND array

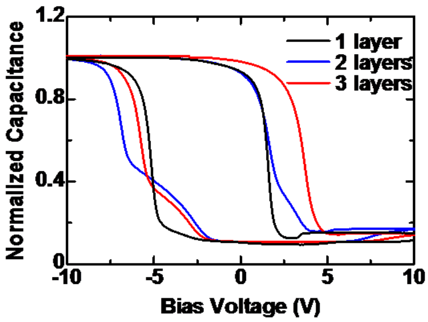

Fig.7. *C-V* characteristics of the MOS capacitors with multi-layered Ni-BND array

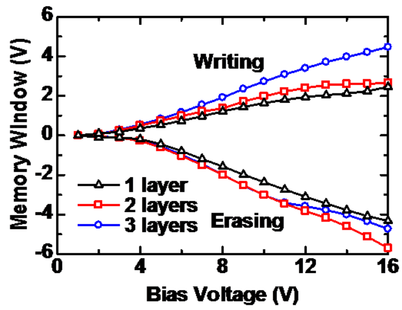

Fig.8. Memory windows of the MOS capacitors with multi-layered Ni-BND array

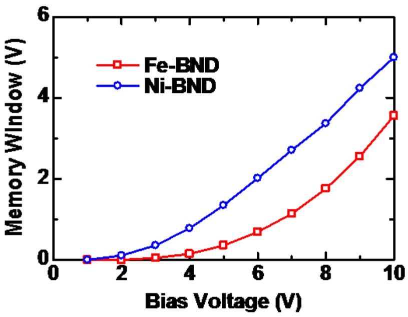

Fig.9. Memory windows of the MOS capacitors with double-layered Fe-BND and Ni-BND array

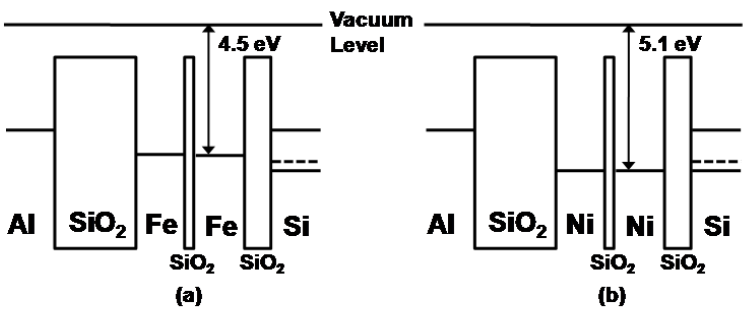

Fig.10. Band diagrams of the MOS capacitors with double-layered (a) Fe-BND array and (b) Ni-BND array