



Article

Internal Resistor Effect of Multilayer-Structured Synaptic Device for Low-Power Operation

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Abstract: A synaptic device with a multilayer structure is proposed to reduce the operating power of neuromorphic computing systems while maintaining a high-density integration. A simple metal-insulator-metal (MIM)-structured multilayer synaptic device is developed using an 8-inch wafer-based and complementary metal-oxide-semiconductor (CMOS) fabrication process. The three types of MIM-structured synaptic devices are compared to assess their effects on reducing the operating power. The obtained results exhibited low-power operation owing to the inserted layers acting as an internal resistor. The modulated operational conductance level and simple MIM structure demonstrate the feasibility of implementing both low-power operation and high-density integration in multilayer synaptic devices.



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Keywords: CMOS compatibility; MIM structure; multilayer synaptic device; low-power operation; inner resistor effect

1. Introduction

The recent exponential growth in unstructured data has led to a significant increase in the amount of data required for efficient processing [1,2]. However, conventional von Neumann computing systems have limitations that result in slow data processing owing to the bottleneck effect caused by the sequential transfer of data between the central processing unit and memory [3–5]. To address this issue, researchers have explored neuromorphic computing systems that use parallel data processing, which enables faster and more energy-efficient processing of large amounts of data [6–8]. To implement this neuromorphic computing system in the hardware, current-based vector–matrix multiplication (VMM) is commonly used via a synaptic device array [9–11]. Because a larger synaptic device array can process more data in parallel, the high-density integration of the synaptic device is necessary. For this purpose, in this research, a simple two-terminal (2T)-based metal–insulator–metal (MIM)-structured memristor which has been studied for memory application is utilized as the synaptic device [12–18].

The 2T-based memristor devices have been investigated, including resistive random-access memory (ReRAM), phase-change memory (PCM) [19], ferroelectric random-access memory (FeRAM) [20,21], and Magnetic random-access memory (MRAM) [22]. Among these memristor devices, ReRAM is the most attractive candidate owing to its simple structure, high-density integration, fast switching speed, and excellent scalability [23–28]. Although the memristor-based synaptic device array can lead to faster parallel data processing using VMM, further research is required to minimize its power consumption. However, ReRAM has been studied for memory application [16,29], research on the device operation mechanism [29–31], and research on ReRAMs composed of materials that are not CMOS-compatible [32,33]. Thus, in this study, a memristor-based 2T synaptic device with a multilayer structure was proposed to reduce the operating power while maintaining high-density integration. Moreover, 8 inch wafer-based CMOS fabrication processes and an

oxide-based W/TaO_x/AlO_x/WO_x/TiN stack were used to assess the feasibility of mass production. The obtained result showed that the AlO_x layer acted as an internal resistor (and barrier layer) without degradation of the synaptic characteristics and exhibited a low-power operation.

2. Materials and Methods

A simple MIM-structured memristor was fabricated to realize the high-density integration of multilayer synaptic devices, as shown in Figure 1. The three types of devices were fabricated to evaluate their effects on reducing the operating power. The W/WO_x/TiN, W/TaO_x/WO_x/TiN, and W/TaO_x/AlO_x/WO_x/TiN stacks were named the single layer, double layer, and triple layer, respectively.

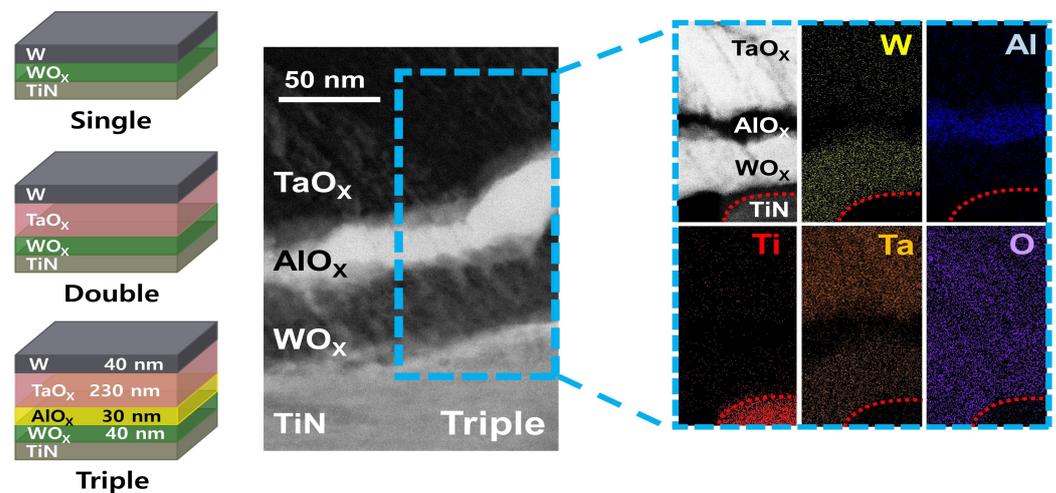


Figure 1. Schematic diagrams of three types of devices with MIM structures; single layer, double layer, and triple layer. Cross-sectional transmission electron microscopy and energy dispersive spectrometry mapping image of the triple layer.

First, a photolithography process was performed to pattern the device. For photolithography, AZ 5214E photoresist (AZ Electronic Materials, Bridgewater, NJ, USA) was applied to the entire wafer using a spin coater. Then, the AZ 300 MIF developer was used for development. A WO_x layer was deposited on the TiN bottom electrode using a typical radio frequency (RF) sputtering system, and a W layer was formed as the top electrode (TE) (called the single layer). Multilayer structures, such as W/TaO_x/WO_x/TiN (called the double layer) and W/TaO_x/AlO_x/WO_x/TiN stack (called the triple layer), were developed and compared to assess their effects on reducing the operating power of the device. All layers were deposited using a sputtering system, and the deposition parameters of each layer are as follows. A 40 nm WO_x channel was deposited by reactive sputtering using a WO₃ target in a 4:1 ratio of Ar and O₂ mixed ambient gas. Then, a 30 nm thick AlO_x layer and a 230 nm thick TaO_x layer were deposited using an Al₂O₃ and Ta₂O₅ target in Ar as the ambient gas. Finally, a 50 nm thick W layer was deposited as the top electrode in ambient Ar gas. WO_x and AlO_x were deposited at a working pressure of 5 mTorr, while TaO_x and W were deposited at 10 m Torr.

Figure 1 shows a cross-sectional transmission electron microscopy (TEM) and energy dispersive spectrometry (EDS) mapping image of the fabricated triple layer. The characteristic X-ray energy of Ta and W elements is 1.709 and 1.774 keV, respectively [34]. Therefore, the W element in the TaO_x region and the Ta element in the WO_x region may overlap. The fabrication processes were based on 8 inch wafer-based CMOS fabrication processes; more details are described in reference [35]. All electrical measurements were conducted using a semiconductor parameter analyzer (HP 4156A) and a pulse generator (Agilent 81110A).

3. Results and Discussion

As mentioned above, the synaptic devices of single, double, and triple layers were fabricated. To confirm the synaptic characteristics of each device, each weight-update curve was measured (Figure 2a–c). The inset of Figure 2a–c show the pulse conditions for potentiation (conductance increase) and depression (conductance decrease). In the single layer, it exhibited resistive switching, which refers to resistance changes from a high-resistance state to a low-resistance state in the negative bias region, and vice versa. When a positive bias is applied to the TE, the oxygen ions of the WO_x layer are migrated to the TE. This migration results in the formation of an induced oxide layer at the interface between the WO_x layer and TE, resulting in decreased conductance. The thickness of the induced oxide layer increased as a continuous positive pulse bias was applied, and thus the conductance was modulated (Figure 2a,d) [36,37]. In contrast, the weight update curve occurs at the opposite polarity for the double and triple layer (Figure 2b,c). The inset of Figure 2b shows the current-voltage (I–V) curve characteristic of the double layer. Gradual resistive switching of the double layer was observed under optimized conditions. The set process in the positive bias and the reset process in the negative bias are observed. Switching behavior occurred in the WO_x layer depending on the mobile oxygen ions between WO_x and TaO_x layers [35,38]. When the positive bias was applied to the TE, the oxygen ions in the WO_x layer moved to the TaO_x layer. Thus, the amount of oxygen vacancies in WO_x increased, resulting in the potentiation process. Conversely, when the negative bias was applied, the oxygen ions that had moved to the TaO_x layer during the potentiation process moved back to the WO_x layer, resulting in the depression process.

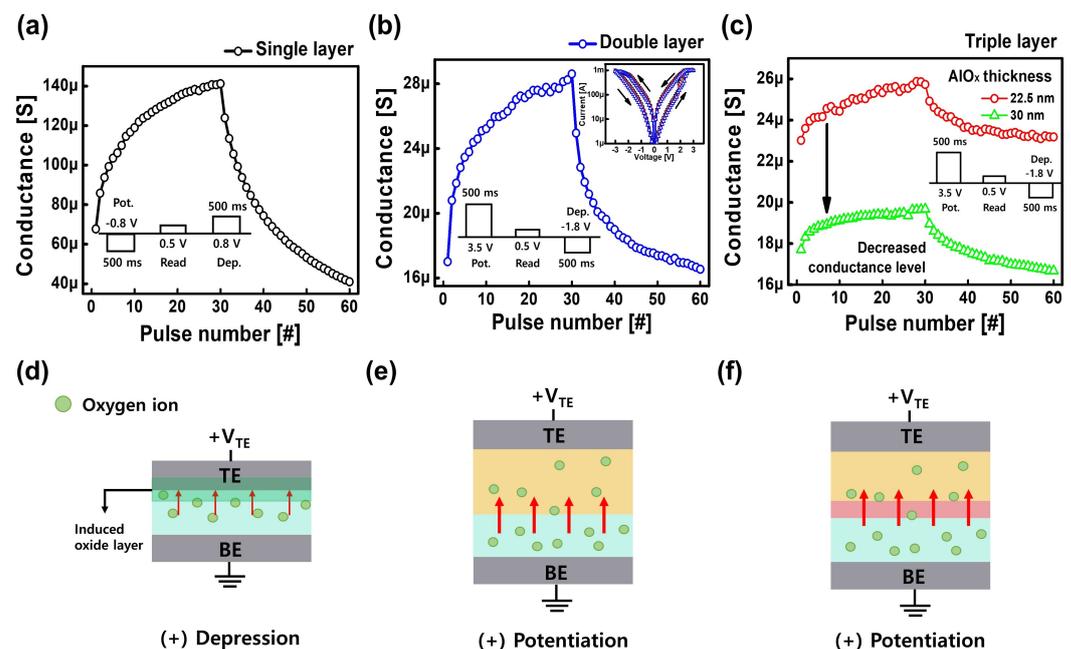


Figure 2. Synaptic characteristics of the weight-update curve in the (a) single, (b) double, and (c) triple layer. The inset shows the optimized pulse amplitude and width (Pot: -0.8 V, 500 ms/Dep: $+0.8$ V, 500 ms for single layer and Pot: $+3.5$ V, 500 ms/Dep: -1.8 V, 500 ms for double and triple layer). (c) Potentiation and depression depend on the thickness of the AlO_x layer in the triple layer. (d–f) Schematic diagram of the operation mechanism in the single, double, and triple layer, respectively.

To achieve synaptic characteristics based on this operating mechanism, the fabrication conditions (such as the Ar: O_2 ratio of the WO_x layer and the working pressure of the TaO_x) were optimized, as shown in Figure 3. A higher initial resistance was observed during the deposition of the WO_x when the Ar: O_2 ratio was increased (Figure 3a). However, resistive switching was only obtained when the ratio of Ar to O_2 was 20:5. This result can be explained in terms of the oxygen vacancy density in the WO_x layer [39] (Figure 3b).

When the Ar:O₂ ratio changed to 20:1, more oxygen vacancies were present in the WO_X layer, resulting in an electrically short state. In contrast, when the Ar:O₂ ratio was 20:10, sufficient oxygen ions were supplied during the deposition of the WO_X. Consequently, an insulating WO_X layer was formed, leading to an electrically insulating behavior.

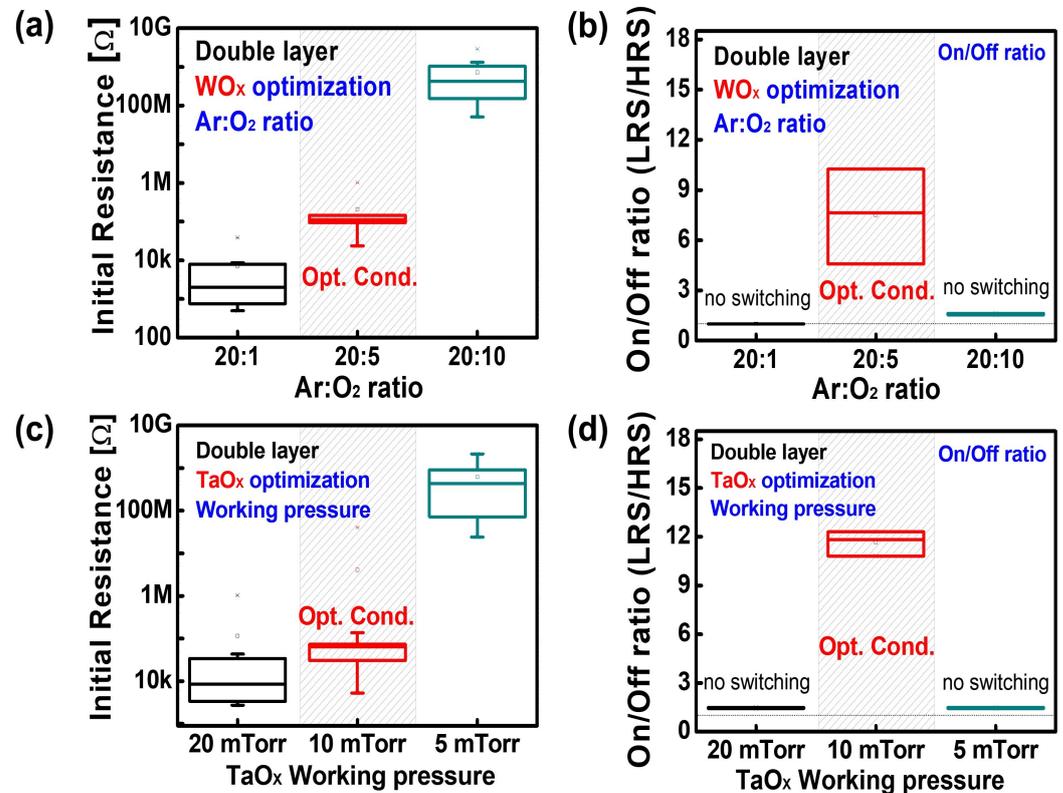


Figure 3. (a) Initial resistance and (b) on/off ratio depending on the Ar:O₂ ratio of the WO_X layer. The resistive switching characteristic appears only under the 20:5 optimized condition. (c) Initial resistances with varying TaO_X layer working pressures. (d) On/off ratio according to the TaO_X layer working pressures. The resistive switching characteristic appears only under 10 mTorr. (Reproduced from Ref. [35] with permission from the Royal Society of Chemistry.)

Based on the optimized WO_X oxygen partial pressure condition, the working pressure of the TaO_X layer was also varied to achieve synaptic characteristics, as shown in Figure 3c,d. When the working pressure was changed from 20 to 10 and 5 mTorr, resistive switching was observed only at 10 mTorr. Considering that a higher working pressure can result in a porous film, deposition at 20 mTorr forms a more porous TaO_X layer [37,40]. Similarly, a denser TaO_X layer was deposited at 5 mTorr. Because the effective area of the interface between the TaO_X and WO_X layers can be increased by higher porosity, more oxygen absorption, resulting in an electrically short state, can occur at 20 mTorr. Additionally, at 5 mTorr, the reduced effective interfacial area and formation of a denser TaO_X layer prevented oxygen absorption. Based on these results, conditions such as an Ar:O₂ ratio of 20:5 and a working pressure of 10 mTorr were selected as the optimal fabrication conditions for the WO_X and TaO_X layers.

The double layer exhibited a lower conductance level than the single layer; however, it was still unacceptably high for the low-power operation of synaptic devices. To further reduce the operating power of the synaptic device, an AlO_X layer was inserted into the interface between the TaO_X and WO_X layers (triple layer). The AlO_X layer was added between the TaO_X and WO_X layers, rather than elsewhere, to obtain the synaptic characteristic. When the AlO_X layer was added to the interface between the WO_X and TiN layers (W/TaO_X/WO_X/AlO_X/TiN), no switching characteristic was observed. The triple layer

has an operating mechanism similar to the double layer. The switching occurs in the WO_X layer according to the mobility of oxygen ions between the WO_X layer and TaO_X layer, as shown in Figure 2f. When the positive bias is applied to the top electrode, oxygen ions in the WO_X layer migrate through the AlO_X layer to the TaO_X layer, causing switching in the WO_X layer. Thus, the potentiation process occurs in which the conductance increases under a positive bias. The conductance level of potentiation and depression decreased with the insertion of the AlO_X layer. The thickness of the AlO_X layer was varied from 22.5 to 30 nm for optimization. The initial resistance increased with increasing AlO_X layer thickness. Owing to the increased initial resistance, the conductance levels of potentiation and depression decreased.

The conductance levels of potentiation and depression were compared in three types of synapse devices. The conductance levels of potentiation and depression decreased with increasing number of layers (Figure 4a). The synaptic characteristics of the devices were verified by normalizing and comparing the potentiation and depression behaviors of the single, double, and triple layers using Equation (1), where G_{max} and G_{min} are the maximum conductance state and minimum conductance state, respectively. The normalized synaptic potentiation and depression behaviors of each device were similar, indicating that the multilayer structure can reduce the operating power without significantly degrading the synaptic characteristics (Figure 4b).

$$G_{normal} = \frac{(G - G_{min})}{(G_{max} - G_{min})} \quad (1)$$

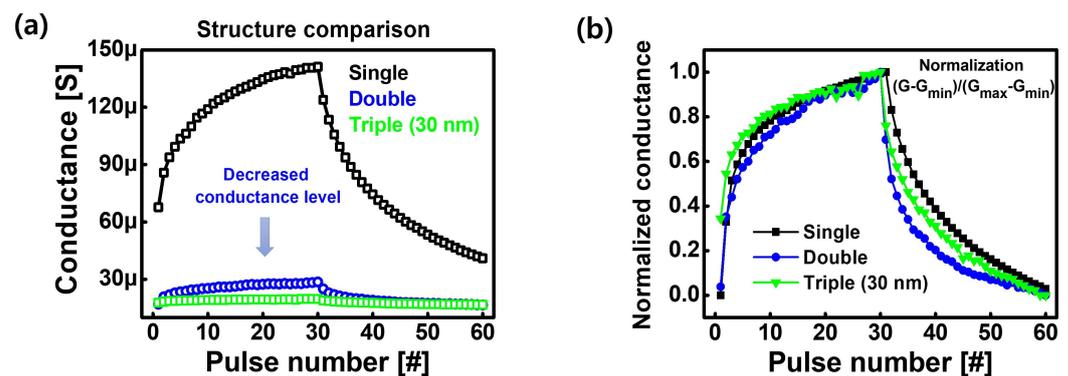


Figure 4. (a) Comparison of the conductance levels of potentiation and depression in the three types of devices. The conductance levels of potentiation and depression decreased with increasing number of layers. (b) Normalized conductance of the single, double, and triple layer devices in potentiation and depression curves. The plot is employed to compare the synaptic characteristics of the devices.

To investigate the role of the inserted AlO_X layer, three cases, namely a double layer, a double layer with an external commercial resistor (200 k Ω), and a triple layer, were compared in Figure 5. Figure 5a compares the double and triple layers, revealing an obvious decrease in the conductance level of the triple layer. As shown in Figure 5b, the conductance of double layer with an external commercial resistor was measured by connecting a 200 k Ω commercial resistor in series through the wiring outside of the double layer device. When the external resistor was connected to the double layer, the conductance level decreased. Compared with the double layer, as shown in Figure 5c, both the triple and double layers with an external resistor exhibited significantly decreased conductance levels. Furthermore, the triple layer exhibited the same operating conductance level as the double layer connected to the external resistor. This result implies that the inserted AlO_X layer can serve as an internal 200 k Ω resistor to efficiently reduce the conductance level.

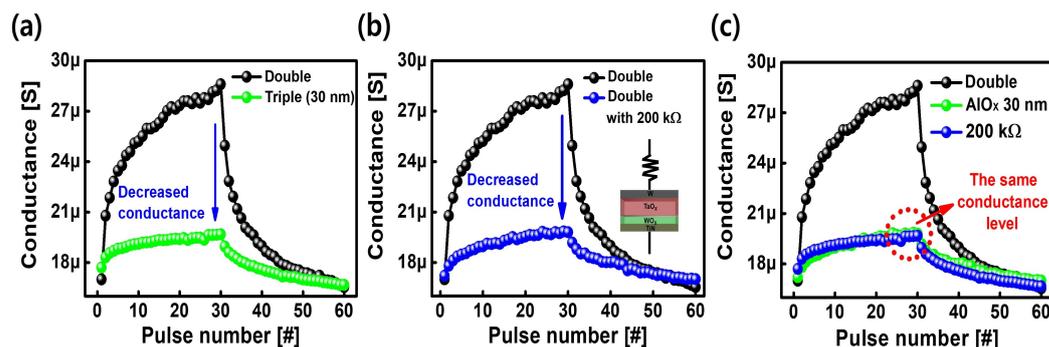


Figure 5. Comparison of the potentiation and depression characteristics of the double layer device with those of (a) a 30 nm-thick AlO_x layer in the triple layer (Triple-30), (b) a double layer with a 200 k Ω resistor (Double-200 k Ω), and (c) both Triple-30 and Double-200 k Ω . The results indicate that the inserted AlO_x layer can serve as an internal 200 k Ω resistor.

In addition, the composition ratio of the WO_x layer, which is a switching layer, was changed compared to the double layer because the AlO_x layer was inserted between the WO_x layer and the TaO_x layer in the triple layer. When the AlO_x layer, which acts as a barrier layer (or shielding layer) [41], was deposited on the WO_x layer, the amount of oxygen ions absorbed from the WO_x layer was reduced. Accordingly, compared with the double layer, the oxygen vacancy density of the WO_x layer of the triple layer decreases. These results were quantitatively analyzed by X-ray photoelectron spectroscopy (XPS) measurements in Figure 6. Figure 6a,b show the XPS analysis spectra of O 1s in the WO_x layer of the double layer and the triple layer, respectively.

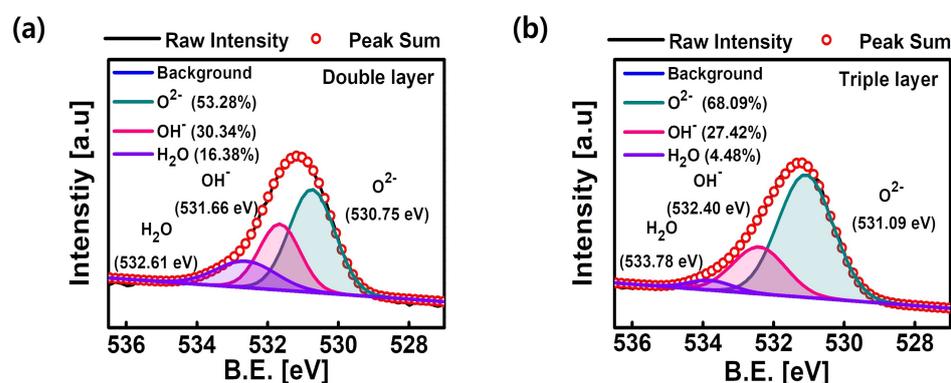


Figure 6. XPS analysis spectra of O 1s in the WO_x layer of (a) the double layer and (b) the triple layer.

The XPS spectrum showed a broad peak, which can be deconvoluted into three individual peaks: the W-O bond peak, oxygen vacancy density, and chemisorbed oxygen species. The green peak of the double layer (530.75 eV) and the triple layer (531.09 eV) can be assigned to the oxygen atoms (O^{2-}) which form W-O bonds. In addition, the violet peaks represent chemisorbed oxygen species (H_2O). Finally, the pink peaks can be assigned to species adsorbed on the surface (OH^- , O^- , or oxygen vacancies); the OH^- groups bond with the metal cations to maintain a charge balance. This implies that the intensity of the OH^- peak indicates oxygen vacancy density [42,43]. The oxygen vacancy density of the double and triple layer are 30.34% and 27.42%, respectively. Therefore, the triple layer has a lower oxygen vacancy density than the double layer. The stoichiometric ratio between tungsten and oxygen can be determined from the composition ratio. In double layer, the tungsten atomic ratio is 30.76% and the oxygen atomic ratio is 69.24%. Thus, the ratio of the tungsten to the oxygen is about 1:2.25 ($\text{WO}_{2.25}$). In the same way, the atomic ratio of tungsten in the triple layer is 29.08% and the atomic ratio of oxygen is 70.92%, so the ratio is 1:2.44 ($\text{WO}_{2.44}$) (Table 1). This indicates that the WO_x of the triple layer contains a smaller number of oxygen vacancies compared to the WO_x of the double layer. As a result, the

AlO_X layer plays the role of $200 \text{ k}\Omega$ because the defect in the switching layer (WO_X layer) decreases the resistance of the AlO_X layer itself. Therefore, the conductance level of the triple layer decreases.

Table 1. Summary of the atomic ratios of W and O of the WO_X layer of the double and triple layers.

| Device | Material | Atomic Ratio (%) | Condition |
|------------------------------------|----------|------------------|---|
| Double layer $\text{WO}_{2.25}$ | W 4f | 30.76 | WO_3 target Ar: $\text{O}_2 = 20:5$ |
| | O 1s | 69.24 | |
| Triple layer $\text{WO}_{2.44}$ | W 4f | 29.08 | |
| | O 1s | 70.92 | |

Owing to the decreased or modulated conductance level, the synaptic device for the neuromorphic system can achieve low power consumption. The power consumption of the single, double, and triple layer was numerically calculated as shown in Figure 7a. When comparing the single and double layer, the power consumption of the double layer was slightly decreased, from $28.24 \mu\text{J}$ to $25.03 \mu\text{J}$. This is because a voltage drop occurred by inserting a TaO_X layer. Thus, a larger pulse amplitude is required for the double layer, and the power consumption was only slightly decreased. However, the power consumption of the triple layer was reduced by 31.2% compared to the double layer (from $25.03 \mu\text{J}$ to $17.22 \mu\text{J}$), with the same pulse width and amplitude. Considering the huge size of the synaptic array in the neuromorphic system, a significant reduction in power consumption can be expected.

Additionally, to verify the influence at the system level, an image recognition simulation consisting of four-layer neural networks was conducted, as shown in Figure 7b–e. The IBM Analog Hardware Acceleration Kit (AIHWKIT), which can simulate devices in real-world applications, is used to simulate training and inference [44]. This provides several device models. We used a “LinearStepDevice” among them. Each parameter required for the simulation was extracted from the measured potentiation/depression weight update curve of the single, double, and triple layer. The neural network was constructed with an input layer of 784 neuron nodes, hidden layer 1 of 256 neuron nodes, hidden layer 2 of 128 neuron nodes, and an output layer of 10 neuron nodes (Figure 7b). A synapse device model was used to connect each neuron node. For the implementation of the deep neural network of Figure 7b at the device level, a synaptic device acting as a weight value can be constructed by a cross-point array [45]. To perform the Multiply and Accumulation operation, the input voltage bias is applied to all row lines, and the output is obtained as a summed current by multiplying the conductance stored at the synaptic devices (Figure 7c). We utilized the Modified National Institute of Standards and Technology (MNIST) dataset (28×28) as an input image. Figure 7d,e show the recognition rate according to the training epoch. The image recognition rates are 85.10%, 71.51%, and 84.11% for a single, double, and triple layer when the numerically ideal case is 92.57%. Even though the triple layer has the lowest power consumption, it exhibited a similar recognition rate to others. This is because the linearity of the weight update curve was not degraded with the addition of the layer compared to the single layer. The image recognition rate reaches about 85%, which is respectable but could be even higher with a wider dynamic range.

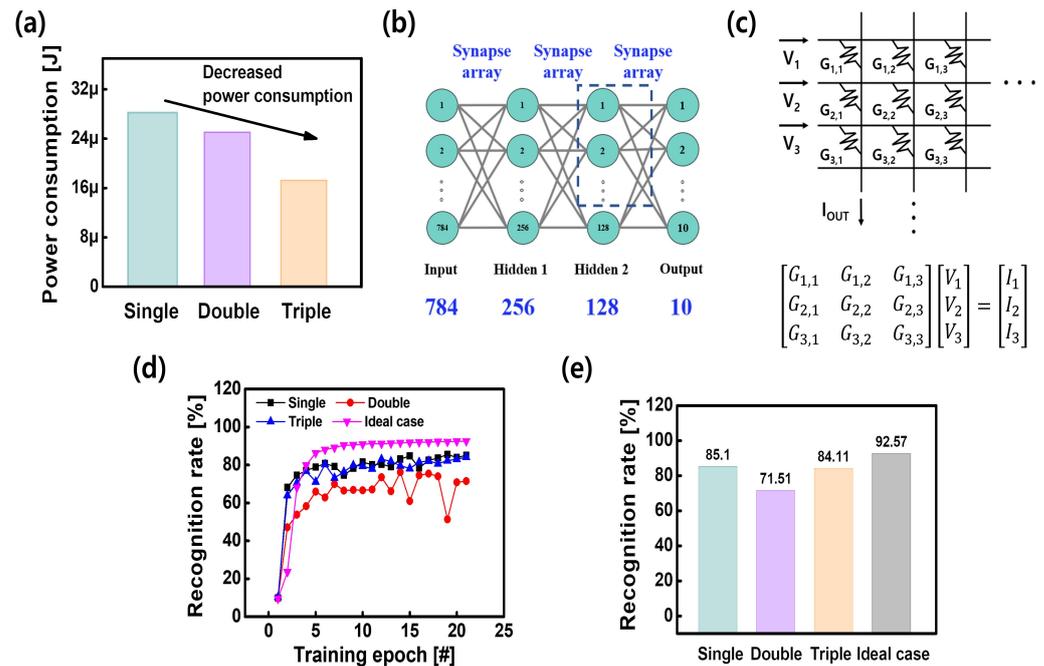


Figure 7. (a) The power consumption of the single, double, and triple layer for the MNIST pattern recognition. (b) Schematic of the neural network; the neural network was constructed with 784 inputs \times 256 first hidden \times 128 second hidden \times 10 output neurons. (c) The crossbar array consists of vertical rows and columns with resistive synaptic devices sandwiched at each cross-point. Recognition rate (d) during and (e) after 20 epochs for single, double, triple layer, and ideal case.

4. Conclusions

In this study, the synaptic device with multilayer MIM-structured synaptic devices suitable for high-density integration and low-power operation were developed using 8 inch wafer-based CMOS fabrication processes. Compared to the double layer, the triple layer demonstrated a low-power operation as the power consumption was reduced by approximately 31%. The synaptic device for neuromorphic systems achieved a low-power consumption due to the reduced or modulated conductance level, because the AlO_x layer inserted in the triple layer not only acts as a barrier layer but also acts as an internal resistor. In addition, the triple layer does not degrade the synaptic characteristics even when the AlO_x layer is added, so the recognition rate shows the undegraded performance of 84.11%. Therefore, the obtained results demonstrate the feasibility of achieving both a low-power operation and high-density integration in multilayer synaptic devices.

Author Contributions: D.L. conceived and directed the research. H.K. and G.H. conducted the experiment. H.K., S.C. and G.H. analyzed the results. H.K. wrote the manuscript. J.W. revised the manuscript. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: The data presented in this study are available on request from the corresponding author.

Conflicts of Interest: The authors declare no conflicts of interest.

References

1. Hammad, K.A.I.; Fakharaldien, M.A.I.; Zain, J.; Majid, M. Big data analysis and storage. In Proceedings of the International Conference on Operations Excellence and Service Engineering, Singapore, 6–9 December 2015; pp. 10–11.
2. Dhanda, N. Big Data Storage and Analysis. *Adv. Data Sci. Anal. Concepts Paradig.* **2022**, *10*, 293–312.
3. Xu, X.; Ding, Y.; Hu, S.X.; Niemier, M.; Cong, J.; Hu, Y.; Shi, Y. Scaling for edge inference of deep neural networks. *Nat. Electron.* **2018**, *1*, 216–222. [[CrossRef](#)]
4. Cai, F.; Correll, J.M.; Lee, S.H.; Lim, Y.; Bothra, V.; Zhang, Z.; Flynn, M.P.; Lu, W.D. A fully integrated reprogrammable memristor–CMOS system for efficient multiply–accumulate operations. *Nat. Electron.* **2019**, *2*, 290–299. [[CrossRef](#)]
5. Tang, J.; Yuan, F.; Shen, X.; Wang, Z.; Rao, M.; He, Y.; Sun, Y.; Li, X.; Zhang, W.; Li, Y.; et al. Bridging biological and artificial neural networks with emerging neuromorphic devices: Fundamentals, progress, and challenges. *Adv. Mater.* **2019**, *31*, 1902761. [[CrossRef](#)] [[PubMed](#)]
6. Mead, C. Neuromorphic electronic systems. *Proc. IEEE* **1990**, *78*, 1629–1636. [[CrossRef](#)]
7. Indiveri, G.; Liu, S.C. Memory and information processing in neuromorphic systems. *Proc. IEEE* **2015**, *103*, 1379–1397. [[CrossRef](#)]
8. Jeong, D.S.; Kim, I.; Ziegler, M.; Kohlstedt, H. Towards artificial neurons and synapses: A materials point of view. *Rsc Adv.* **2013**, *3*, 3169–3183. [[CrossRef](#)]
9. Marinella, M.J.; Agarwal, S.; Hsia, A.; Richter, I.; Jacobs-Gedrim, R.; Niroula, J.; Plimpton, S.J.; Ipek, E.; James, C.D. Multiscale co-design analysis of energy, latency, area, and accuracy of a ReRAM analog neural training accelerator. *IEEE J. Emerg. Sel. Top. Circuits Syst.* **2018**, *8*, 86–101. [[CrossRef](#)]
10. Kim, S.; Baek, M.H.; Hwang, S.; Jang, T.; Park, K.; Park, B.G. A novel vector-matrix multiplication (VMM) architecture based on NAND memory array. *J. Semicond. Technol. Sci.* **2020**, *20*, 242–248. [[CrossRef](#)]
11. Han, G.; Lee, C.; Lee, J.E.; Seo, J.; Kim, M.; Song, Y.; Seo, Y.H.; Lee, D. Alternative negative weight for simpler hardware implementation of synapse device based neuromorphic system. *Sci. Rep.* **2021**, *11*, 23198. [[CrossRef](#)]
12. Mohammad, B.; Jaoude, M.A.; Kumar, V.; Al Homouz, D.M.; Nahla, H.A.; Al-Qutayri, M.; Christoforou, N. State of the art of metal oxide memristor devices. *Nanotechnol. Rev.* **2016**, *5*, 311–329. [[CrossRef](#)]
13. Zhu, K.; Mahmoodi, M.R.; Fahimi, Z.; Xiao, Y.; Wang, T.; Bukvišová, K.; Kolíbal, M.; Roldan, J.B.; Perez, D.; Aguirre, F.; et al. Memristors with Initial Low-Resistive State for Efficient Neuromorphic Systems. *Adv. Intell. Syst.* **2022**, *4*, 2200001. [[CrossRef](#)]
14. Saludes-Tapia, M.; Campabadal, F.; Miranda, E.A.; Gonzalez, M.B. Impact of the W Etching Process on the Resistive Switching Properties of Tin/Ti/HfO₂/W Memristors. *Solid-State Electron.* **2023**, *207*, 108718. [[CrossRef](#)]
15. Mohanty, S.K.; Panda, D.; Reddy, K.P.K.; Lee, P.T.; Wu, C.H.; Chang, K.M. Uniform resistive switching and highly stable synaptic characteristics of HfO_x sandwiched TaO_x-based memristor for neuromorphic system. *Ceram. Int.* **2023**, *49*, 16909–16917. [[CrossRef](#)]
16. Basnet, P.; Anderson, E.C.; Athena, F.F.; Chakrabarti, B.; West, M.P.; Vogel, E.M. Asymmetric Resistive Switching of Bilayer HfO_x/AlO_y and AlO_y/HfO_x Memristors: The Oxide Layer Characteristics and Performance Optimization for Digital Set and Analog Reset Switching. *Acs Appl. Electron. Mater.* **2023**, *5*, 1859–1865. [[CrossRef](#)]
17. Zhu, S.; Sun, B.; Zhou, G.; Guo, T.; Ke, C.; Chen, Y.; Yang, F.; Zhang, Y.; Shao, J.; Zhao, Y. In-Depth Physical Mechanism Analysis and Wearable Applications of HfO_x-Based Flexible Memristors. *Acs Appl. Mater. Interfaces* **2023**, *15*, 5420–5431. [[CrossRef](#)] [[PubMed](#)]
18. Bature, U.I.; Nawari, I.M.; Khir, M.H.M.; Zahoor, F.; Hashwan, S.S.B.; Algamili, A.S.; Abbas, H. Analysis of thermodynamic resistive switching in ZnO-based RRAM device. *Phys. Scr.* **2023**, *98*, 035020. [[CrossRef](#)]
19. Kim, S.; Ishii, M.; Lewis, S.; Perri, T.; BrightSky, M.; Kim, W.; Jordan, R.; Burr, G.W.; Sosa, N.; Ray, A.; et al. NVM neuromorphic core with 64k-cell (256-by-256) phase change memory synaptic array with on-chip neuron circuits for continuous in-situ learning. In Proceedings of the 2015 IEEE International Electron Devices Meeting (IEDM), Washington, DC, USA, 7–9 December 2015; pp. 17.1.1–17.1.4.
20. Oh, S.; Kim, T.; Kwak, M.; Song, J.; Woo, J.; Jeon, S.; Yoo, I.K.; Hwang, H. HfZrO_x-based ferroelectric synapse device with 32 levels of conductance states for neuromorphic applications. *IEEE Electron Device Lett.* **2017**, *38*, 732–735. [[CrossRef](#)]
21. Grenouillet, L.; Francois, T.; Coignus, J.; Vaxelaire, N.; Carabasse, C.; Triozon, F.; Richter, C.; Schroeder, U.; Nowak, E. Performance assessment of BEOL-integrated HfO₂-based ferroelectric capacitors for FeRAM memory arrays. In Proceedings of the 2020 IEEE Silicon Nanoelectronics Workshop (SNW), Honolulu, HI, USA, 13–14 June 2020; pp. 5–6.
22. Rahaman, S.Z.; Chang, Y.J.; Hsin, Y.C.; Yang, S.Y.; Lee, H.H.; Wang, I.J.; Chen, G.L.; Su, Y.H.; Wei, J.H.; Sheu, S.S.; et al. Process-induced magnetic tunnel junction damage and its recovery for the development of spin-orbit torque magnetic random access memory. *J. Magn. Magn. Mater.* **2023**, *565*, 170296. [[CrossRef](#)]
23. Waser, R.; Dittmann, R.; Staikov, G.; Szot, K. Redox-based resistive switching memories—nanoionic mechanisms, prospects, and challenges. *Adv. Mater.* **2009**, *21*, 2632–2663. [[CrossRef](#)]
24. Chand, U.; Huang, C.Y.; Kumar, D.; Tseng, T.Y. Metal induced crystallized poly-Si-based conductive bridge resistive switching memory device with one transistor and one resistor architecture. *Appl. Phys. Lett.* **2015**, *107*, 203502. [[CrossRef](#)]
25. Baek, I.; Kim, D.; Lee, M.; Kim, H.J.; Yim, E.; Lee, M.; Lee, J.; Ahn, S.; Seo, S.; Lee, J.; et al. Multi-layer cross-point binary oxide resistive memory (OxRRAM) for post-NAND storage application. In Proceedings of the IEEE International Electron Devices Meeting, 2005, IEDM Technical Digest, Washington, DC, USA, 5 December 2005; pp. 750–753.
26. Sawa, A. Resistive switching in transition metal oxides. *Mater. Today* **2008**, *11*, 28–36. [[CrossRef](#)]

27. Luo, Q.; Xu, X.; Gong, T.; Lv, H.; Dong, D.; Ma, H.; Yuan, P.; Gao, J.; Liu, J.; Yu, Z.; et al. 8-Layers 3D vertical RRAM with excellent scalability towards storage class memory applications. In Proceedings of the 2017 IEEE International Electron Devices Meeting (IEDM), San Francisco, CA, USA, 2–6 December 2017; pp. 2.7.1–2.7.4. [\[CrossRef\]](#)
28. Luo, Q.; Zhang, X.; Yu, J.; Wang, W.; Gong, T.; Xu, X.; Yin, J.; Yuan, P.; Tai, L.; Dong, D.; et al. Memory Switching and Threshold Switching in a 3D Nanoscaled NbOX System. *IEEE Electron Device Lett.* **2019**, *40*, 718–721. [\[CrossRef\]](#)
29. Zhang, W.; Guo, Z.; Dai, Y.; Lei, J.; Wang, J.; Hu, F. Effects of stacking sequence and top electrode configuration on switching behaviors in ZnO-HfO₂ hybrid resistive memories. *Ceram. Int.* **2023**, *49*, 35973–35981. [\[CrossRef\]](#)
30. Kim, Y.; Jo, K.J.; Oh, J.S.; Yang, C.W. Bipolar Resistive Switching Characteristics of Ta/TaxMnyOz/Pt Structure for ReRAM Application with Large Resistance Window. *Electron. Mater. Lett.* **2023**, *20*, 26–32. [\[CrossRef\]](#)
31. Das, O.P.; Pandey, S.K. Optical, Compositional and Electrical Properties of Transparent MgO Thin Film for ReRAM Devices. *Proc. J. Phys. Conf. Ser.* **2023**, *2426*, 012031. [\[CrossRef\]](#)
32. Moazzeni, A.; Kordrostami, Z. Switching characteristic of fabricated nonvolatile bipolar resistive switching memory (ReRAM) using PEDOT: PSS/GO. *Solid-State Electron.* **2022**, *188*, 108208. [\[CrossRef\]](#)
33. Lodhi, A.; Saini, S.; Dwivedi, A.; Khandelwal, A.; Tiwari, S.P. Bipolar resistive switching properties of TiO_x/graphene oxide doped PVP based bilayer ReRAM. *J. Micromech. Microeng.* **2022**, *32*, 044001. [\[CrossRef\]](#)
34. Mu, S.; Stowe, D. Accurate Elemental Mapping of Semiconductor Devices Using EDS–Deconvolving Overlapping Peaks. *Microsc. Microanal.* **2023**, *29*, 107–108. [\[CrossRef\]](#)
35. Han, G.; Seo, J.; Kim, H.; Lee, D. Role of the electrolyte layer in CMOS-compatible and oxide-based vertical three-terminal ECRAM. *J. Mater. Chem.* **2023**, *11*, 5167–5173. [\[CrossRef\]](#)
36. Kanegami, N.; Nishi, Y.; Kimoto, T. Unique resistive switching phenomena exhibiting both filament-type and interface-type switching in Ti/PrO₃·3CaO·3MnO₃- δ /Pt ReRAM cells. *Appl. Phys. Lett.* **2020**, *116*, 013501. [\[CrossRef\]](#)
37. Park, J.; Lee, C.; Kwak, M.; Chekol, S.A.; Lim, S.; Kim, M.; Woo, J.; Hwang, H.; Lee, D. Microstructural engineering in interface-type synapse device for enhancing linear and symmetric conductance changes. *Nanotechnology* **2019**, *30*, 305202. [\[CrossRef\]](#) [\[PubMed\]](#)
38. Pershin, Y.V.; Martinez-Rincon, J.; Di Ventra, M. Memory circuit elements: From systems to applications. *J. Comput. Theor. Nanosci.* **2011**, *8*, 441–448. [\[CrossRef\]](#)
39. Rudrapal, K.; Biswas, M.; Jana, B.; Adyam, V.; Chaudhuri, A.R. Tuning resistive switching properties of WO₃-x-memristors by oxygen vacancy engineering for neuromorphic and memory storage applications. *J. Phys. Appl. Phys.* **2023**, *56*, 205302. [\[CrossRef\]](#)
40. Lee, C.; Choi, W.; Kwak, M.; Kim, S.; Hwang, H. Impact of electrolyte density on synaptic characteristics of oxygen-based ionic synaptic transistor. *Appl. Phys. Lett.* **2021**, *119*, 103503. [\[CrossRef\]](#)
41. Lee, C.; Choi, W.; Kwak, M.; Kim, S.; Hwang, H. Excellent synapse characteristics of 50 nm vertical transistor with WO_x channel for high density neuromorphic system. In Proceedings of the 2021 Symposium on VLSI Technology, Kyoto, Japan, 13–19 June 2021; pp. 1–2.
42. Mazur, M.; Wojcieszak, D.; Wiatrowski, A.; Kaczmarek, D.; Lubańska, A.; Domaradzki, J.; Mazur, P.; Kalisz, M. Analysis of amorphous tungsten oxide thin films deposited by magnetron sputtering for application in transparent electronics. *Appl. Surf. Sci.* **2021**, *570*, 151151. [\[CrossRef\]](#)
43. Rahimnejad, S.; He, J.H.; Chen, W.; Wu, K.; Xu, G.Q. Tuning the electronic and structural properties of WO₃ nanocrystals by varying transition metal tungstate precursors. *RSC Adv.* **2014**, *4*, 62423–62429. [\[CrossRef\]](#)
44. Rasch, M.J.; Moreda, D.; Gokmen, T.; Le Gallo, M.; Carta, F.; Goldberg, C.; El Maghraoui, K.; Sebastian, A.; Narayanan, V. A flexible and fast PyTorch toolkit for simulating training and inference on analog crossbar arrays. In Proceedings of the 2021 IEEE 3rd International Conference on Artificial Intelligence Circuits and Systems (AICAS), Washington, DC, USA, 6–9 June 2021; pp. 1–4.
45. Burr, G.W.; Shelby, R.M.; Sebastian, A.; Kim, S.; Kim, S.; Sidler, S.; Virwani, K.; Ishii, M.; Narayanan, P.; Fumarola, A.; et al. Neuromorphic computing using non-volatile memory. *Adv. Physics X* **2017**, *2*, 89–124. [\[CrossRef\]](#)

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