Amorphous Metal Oxide Bilayers to Avoid Sneak-Path Currents for High-Density Resistive Memory Arrays

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Resistance switching devices are potential candidates for future memory applications. However, sneak-path current issues in crossbar array cause energy and heat dissipation, and it requires additional circuitry for mitigation. To address this, we introduce a hybrid device with functional oxide as bilayer of amorphous strontium titanium oxide (α-STO) and vanadium oxide (α-VOx). High-performance resistive switching material α-STO is coupled with α-VOx having fast apolar threshold switching. The hybrid device gives performance equivalent to conventional one resistance switch and one selector architecture with stability of 6000 endurance cycles. Non-linearity factor of the hybrid device is 4.8, which is slightly higher than only α-STO device of 4.7. Using I–V measurements, readout margin is calculated, which suggests integration into 10⁴ × 10⁶ array when 10% readout margin is considered at switching ratio of 8. Further, the selector effect using non-zero-crossing property of memristors, which reveals six times reduction in OFF current in the hybrid device, is quantified. Compositional analysis along the device cross-section to understand elemental distribution and interface effects of two unique, multifaceted oxides is presented. As such, in the hybrid structure non-linearity factor remains nearly constant, while the absolute value of non-zero-crossing current is reduced six times, which can reduce the overall sneak current in crossbars.

1. Introduction

Digital memory has become indispensable for daily life; however, significant concerns exist due to exponentially increasing demand for small size, high speed, high storage capacity, and low power for next-generation memory devices. Resistive random-access memory (RRAM) is increasingly being used as a building block for novel memory and logic architectures as a next-generation memory device due to its ability to be designed in high-density 3D crossbar arrays. Even while RRAM has excellent advantages for use as digital memories, the expected outcome is not yet achieved due to sneak-path or leakage current problems. It is a critical issue when constructing the crossbar structures, limiting their reliability, storage capacity, and energy efficiency.

To overcome the sneak-path problem, multiple approaches have been reported including different reading schemes, unfolded architecture, three-terminal memristor gating, complementary resistive switch (CRS), using memristor nonlinearity, alternating current (AC) sensing, transistor gating, and diode gating or use of a selection device. Among these, different reading schemes, unfolded architecture, three-terminal memristor gating, and CRS increase the memory operation complexity. AC sensing technique needs additional AC input and sensing equipment, which adds extra complexity to the system. A very useful property—memristor non-linearity—consistently reduces leakage current with high factor; however, it is only applicable to resistive switching materials with high nonlinearity factor. Transistor and diode gating or use of selection device strategies would solve the sneak-path problem. However, transistor gating will prevent high density of crossbars as the size of gating transistor is larger than the resistance switch. One-selector and one-resistor switch (1S1R) is an ideal strategy to eliminate the sneak paths; however, introducing system delays and increasing fabrication and biasing complexity have been reported.

For the class of nonvolatile bipolar resistive switching (BRS) devices, an ideal selector should be fast to avoid delays and should have nonlinear and symmetric current–voltage (I–V) characteristics. Among the possible materials for selectors,
vanadium oxide has been reported for high switching speeds in the picosecond domain.\textsuperscript{[19]} It shows apolar or symmetric, stable, and nonlinear threshold switching (TS) characteristics.\textsuperscript{[20–24]} That being so, the selection properties of vanadium oxide have been reported along with a few resistive switching devices based on hafnium oxide (HfO\textsubscript{2}), silicon dioxide (SiO\textsubscript{2}), and functional bilayer of ZnO/HfO\textsubscript{2}~\textsuperscript{[21–22,25–27]} However, its performance is not yet tested in conjunction with a well-known resistive switching material—strontium titanium oxide (STO). STO is a widely researched material in the field of RRAM having excellent memory properties,\textsuperscript{[28–27]} with a demonstrated ability to realize transparent\textsuperscript{[30]} and flexible\textsuperscript{[38]} crossbar arrays.

In this work, we present a hybrid cross-point device (which can be easily scaled up to crossbar arrays) with a combinatorial arrangement of a bilayer of amorphous strontium titanium oxide (a-STO) and amorphous vanadium oxide (a-VO\textsubscript{x}) as a functional oxide, unlike conventional series connection of a selector and a resistor switch (supporting information Figure S4, Supporting Information). This arrangement simplifies the fabrication process and biasing complexity of 1S1R architecture. The choice of amorphous materials further gives an edge in fabrication by allowing room temperature processing and complementary metal–oxide–semiconductor (CMOS) back-end integration compatibility. The hybrid bilayer device gives a performance equivalent to the 1S1R structure without compromising the high density on a single chip. We present complete electrical characterizations to understand its memory performance including electroforming, switching repeatability, retention, and endurance. We calculated the readout margin (RM) from the \textit{I–V} measurements and suggest the potential array size of the crossbar based on the proposed hybrid device. We further compare the performance of a hybrid device with only a-STO-based memory device. We discuss the origins of sneak-path current and thereby present a quantitative analysis method to understand the selector effect using nonzero-crossing characteristics. Finally, we provide insights into switching mechanism along with a simple elemental analysis of the hybrid device using achieved electrical results and compositional characterizations.

2. Results and Discussion

We present a cross-point device structure with 6 \textmu m × 6 \textmu m electrode overlap and 25 \textmu m × 25 \textmu m oxide patch. The functional oxide layer consists of a bilayer of a-STO–a-VO\textsubscript{x} (80–120 nm) with Pt/Ti (120/20 nm) as top electrode (TE) and Pt (15 nm) as bottom electrode (BE). Electrical characterizations, compositional analysis, and insights into the possible switching mechanism are presented in this section.

2.1. Electrical Characteristics of a Hybrid Device Based on Bilayer of a-STO–a-VO\textsubscript{x} as Functional Oxides

A hybrid device in its pristine state exhibits a resistance of \textasciitilde 80 M\Omega. All the electrical measurements are carried out with BE biased and TE grounded, as shown in Figure 1. To initiate BRS, an irreversible electroforming sweep is required. A typical electroforming curve is shown in Figure 1a. The electroforming voltage ranges from 6 to 10 V and postelectroforming resistance is in the range of 80–100 k\Omega.

After electroforming, BRS is achieved. A typical BRS curve is shown in Figure 1b’s inset. A single cell is tested for over 10\textsuperscript{5} consecutive voltage sweeps, as shown in Figure 1b. With a higher number of consecutive sweeps, a shift in switching curve or threshold voltage is observed. As a-STO has been reported to be extremely stable, this shift in the curve might be mainly due to the Joule heating effects in a-VO\textsubscript{x}.\textsuperscript{[23,34]} Devices are tested for a retention of over 10\textsuperscript{4} s at read voltage of 1 V to verify its nonvolatile nature, as shown in Figure 1c. After this, resistance of the low resistance state (LRS) slowly starts increasing. However, the retention for 10\textsuperscript{4} s confirms that a hybrid device of volatile a-VO\textsubscript{x} and nonvolatile a-STO retains the nonvolatile BRS characteristics of a-STO. Devices are further tested for over 6000 endurance cycles, as shown in Figure 1d. The stability of hybrid device proposed here is significantly (more than 10 times) better than the previously reported conventional 1S1R structures based on vanadium oxide as a selector.\textsuperscript{[21,39]}

Further, to confirm the consistency of the hybrid device, multiple devices were tested and the statistical data for 17 devices are shown in Figure 1e,f. Figure 1e shows the distribution of switching voltage, read voltage, and switching ratio, whereas Figure 1f shows the mean value and standard deviation for the set, reset, read voltage, and switching ratio. From Figure 1f, the mean value for reset voltage is \textasciitilde 2 V, for set voltage it’s 2 V, read voltage 1 V, and the switching ratio is 8 V.

2.2. Comparison of a-STO, a-VO\textsubscript{x} and a-STO–a-VO\textsubscript{x}-Based Hybrid Devices

In this section, we compare resistive switching performance of the hybrid device with individual a-STO and a-VO\textsubscript{x} devices, as shown in Figure 2. Typical electroforming and resistive switching curves are shown in Figure 2a,b for a-STO, Figure 2c,d for a-VO\textsubscript{x}, and Figure 2e,f for the hybrid device. In Figure 2, column one shows typical electroforming curves, whereas column two shows typical resistive switching curves. Electroforming voltages for all the three devices are in the same range of 6–8 V, as shown in Figure 2a,c,e, and the switching voltage range of 1.6–2.5 V is also similar for all the three devices, as shown in Figure 2b,d,f. Resistance switching characteristics are nonvolatile and bipolar in nature for a-STO and the hybrid device, whereas it is volatile and apolar for a-VO\textsubscript{x} device. By comparing Figure 2b and Figure 2d, it can be noticed that unnecessary high current in a-STO-based devices at lower voltages is cancelled in the hybrid device due to the incorporation of a-VO\textsubscript{x} acting as a selector. Observation of LRS current (marked as 3 in magenta) in a-STO (Figure 2b) and in hybrid device (Figure 2f) clarifies the reduction of current at lower voltage values. Further, the ratio of currents at the read voltage and half-read voltage provides the nonlinearity factor, as shown in Figure 2b,d,f. The nonlinearity factor is 4.7 for a-STO, 23.12 for a-VO\textsubscript{x}, and 4.8 for the hybrid device. Higher nonlinearity ratios have been reported earlier.\textsuperscript{[39]} However, please note that the nonlinearity ratio was increased slightly after incorporating a-VO\textsubscript{x} as a selecting bilayer in functional oxides. In future, this ratio can be further improved by tailoring the oxide stoichiometry, deposition conditions, and
This proves that the resistive switching characteristics of the hybrid device are clearly a combination of its individual counterparts with stable and repeatable nonvolatile resistive switching of $a$-STO and TS selector-like behavior of $a$-VO$_x$, while keeping the nonlinearity factor nearly same.

It has been reported by Valov et al.\[40\] that nonequilibrium ON and OFF states exist in practical RRAM devices due to the electromotive force (EMF) in electrochemical systems such as resistive switches. This EMF results in the nonzero-crossing hysteresis loop of memristors.\[40\] We believe that this nonzero-crossing current is the primary cause of sneak-path current in crossbar arrays. Further, origin of leakage current or EMF can be attributed to three effects: 1) Nernst potential which is the potential difference at three interfacial regions, namely, Ti/$a$-STO, $a$-STO/$a$-VO$_x$, and $a$-VO$_x$/Pt;\[41\] 2) diffusion potential which is the potential difference generated due to the diffusion of both oxygen vacancies and vanadium ions; and 3) Gibbs–Thompson effect. This contributing factor is due to different surface free energies of macrosized electrodes, nanosized filament in two different regions $a$-STO and $a$-VO$_x$, and presence of localized VO$_2$ crystal islands within or near the filament.\[23\] Further investigations would be required to ascertain this suggestion and confirm the origin of leakage current using nanostructural analysis in future.

Here, we compare two nonzero-crossing properties—one the OFF voltage current after set and reset cycles and second, the memristive crossing voltage of $a$-STO, $a$-VO$_x$, and $a$-STO/$a$-VO$_x$ devices to understand the quantitative selector effect, as shown in Figure 3.

OFF voltage currents after set and reset cycles are considered separately to understand the quantitative leakage current in each state. After the set cycle, OFF voltage current is $-171 \text{ nA}$ for...
Figure 2. Comparison of resistive switching performance of $a$-VO$_x$ and $a$-STO-based hybrid cross-point device with individual components. a,c,e) Typical electroforming curves with insets showing cross section of corresponding devices. b,d,f) Typical resistive switching curves. (a,b) is for $a$-STO, (c,d) is for $a$-VO$_x$, and (e,f) is for a $a$-STO-$a$-VO$_x$ hybrid device. f) The read voltage (1 V) and half-read voltage (0.5 V) in LRS are marked in (b), (d), and (f) graphs. The nonlinearity ratios for $a$-STO, $a$-VO$_x$, and hybrid devices are 4.7, 23.12, and 4.8, respectively.

Figure 3. Comparative analysis of nonzero-crossing characteristics: Typical resistive switching curves zoomed near origin point for a) $a$-STO, b) $a$-VO$_x$, and c) $a$-STO-$a$-VO$_x$-based cross-point devices. The analysis shows $6\times$ reduction in OFF voltage current after set cycle and $4\times$ reduction in nonzero-crossing voltage in the hybrid device.
a-STO (Figure 3a), which is reduced to −28 nA (Figure 3c) in the hybrid device, which is six times less due to the selector effect of a-VOx. Resistance switching in a-VOx devices is volatile and does not have set or reset states. However, OFF voltage currents after both positive and negative schemes are 3.4 and 3.2 nA, as shown in Figure 3b.

After the reset cycle, the OFF voltage current of 14.3 nA in the hybrid device is slightly higher compared with the a-STO device with 7 nA. However, when the values of set and reset currents in Figure 3a are compared, it should be accepted that the majority of leakage current is generated in the set cycle due to the formed filament and the LRS, which is significantly reduced by the incorporation of selecting second layer. In the reset cycle, we attribute the opposite behavior to the Gibbs–Thompson effect. Gibbs–Thompson effect is directly proportional to the molar volume of the dielectric functional layer, which varies with the thickness of films. As such, it is obvious that the amount of leakage current would be higher for thicker functional oxide. For this reason, non-zero-crossing current for the bilayer hybrid device (having twice the thickness of functional layer) is slightly higher than only a-STO device in the RESET cycle. For similar reasons, we suggest that the observed leakage current in the RESET cycle can be reduced by thinner functional oxide bilayer. This claim is supported by the results shown in Figure S3, Supporting Information, that present preliminary resistive switching characteristics of the hybrid device with 40 nm: 40 nm bilayer of a-STO and a-VOx.

Improvement in sneak-path current can also be presented by considering the non-zero-crossing voltage values. In the a-STO-based device, crossing voltage is 7.6 mV due to nonequilibrium, as shown in Figure 3a. To establish the equilibrium, gradually, an equivalent amount corresponding current would leak in crossbar arrays. To avoid this, same magnitude of voltage of reverse polarity is needed to bring about zero-crossing requiring, which in turn requires higher power. In the volatile a-VOx, crossing voltage is zero, as shown in Figure 3b. In the hybrid device (Figure 3c), the crossing voltage is reduced to 1.8 mV, which is nearly four times less compared to the a-STO-based device.

The comparison of a-STO and hybrid device suggests that change in nonlinearity factor after incorporation of a-VOx is minimal. Further, as per existing theories and models, nearly constant nonlinearity factor means minimal improvement in RM and size of crossbar arrays. However, from Figure 3, there is six times reduction in the OFF current, which is a significant change. The effect of the absolute value of OFF current on the RM and array size is an unexplored area. The effect of OFF current on the overall sneak-path current is not clear yet and has to be investigated further in future.

To estimate the scalability of crossbar array of the hybrid device based on a-STO and a-VOx bilayer, the RM is calculated using the superposition theory. The RM for the hybrid device at the switching ratio of 8 is 47.8%. Using the device parameters extracted from Figure 2b, the RM is also obtained as a function of number of words depending on the minimum, maximum, and mean switching ratios shown in Figure 1f. We used a read scheme of + 1V_read and −1V_read to access the selected cell and grounded nonaccessed word/bit lines, which is the worst case. The graph of RM versus number of words is shown in Figure 4a. It suggests that at 10% RM, the potential array size for our hybrid device can be 10^4 × 10^4 at the switching ratio of 8, which is comparable with the conventional 15J architectures. Further, array size for only a-VOx device (at switching ratio 76.4) is three times higher than the hybrid device (at switching ratio 8) when RM of 10% is considered. This highlights that although only selector device based on a-VOx suggests higher device density, practically, in combination with resistive switching device, achievable array sizes can be lower.

A significantly higher switching ratio of ≈10^4 has been reported for a-STO devices. The primary reason for achieving very-low switching ratio in this work compared with earlier reports is the quality and thickness of a-STO film. However, from the presented study, it is clear that the change in the switching ratio of the hybrid device is minimal compared with a-STO device. This highlights the potential to improve switching ratio of the hybrid device up to ≈10^5 by tailoring the properties of a-STO film in future. To estimate the best possible array size for presented hybrid device structure, RM versus number of words curve at a switching ratio of 10^4 is included in Figure 4a, which suggests five time increase in the array size compared with the current hybrid device.

Figure 4b shows the area dependence of the switching voltages for hybrid device. The results show that the switching voltages for different-sized devices are within a range from ±2 to ±4 V.

Figure 4. Scalability analysis. a) Relationship between RM (a percentage of applied wordline bias), crossbar memory array size, and switching ratio of the hybrid device. 10% RM is the assumed criteria for read operation (worst case). Straight line graphs correspond to the hybrid device structure, whereas dashed line represents only a-VOx device. b) SET and RESET voltages as a function of device area. The graph shows that the switching voltages for different-sized devices are within a range from ±2 to ±4 V.
2.3. Compositional Analysis and Interface Effects

Electrical analysis proves that the bilayer of $a$-STO and $a$-VO$_x$ as a functional oxide in cross-point structure gives performance equivalent to 1S1R structure. This work suggests that instead of connecting a selector and a memory resistor separately in series, hybrid device structure is advantageous. However, before practical application, it is important to understand the elemental distribution and interface effects along the stacks of the pristine device.

We used electron energy loss spectroscopy (EELS) and transmission electron microscopy (TEM) characterization to understand the elemental distribution, interface effects, and evolution of oxidation states of Ti and V across the hybrid device cross section, as shown in Figure 5. To achieve the comparative elemental distribution, EELS line scan is collected for a broader distribution window of 140 eV to cover titanium (Ti), vanadium (V), and oxygen (O) spectra, as shown in scanning transmission electron micrograph in Figure 5a. The background-subtracted EELS spectra across the TE (Ti), Ti/$a$-STO interface, bulk $a$-STO, $a$-STO/$a$-VO$_x$ interface, bulk $a$-VO$_x$, and Pt are shown in Figure 5b, highlighting the elemental distribution at these regions.

Starting from the TE, Ti has two peaks $L_1$ ($e_g$) at 458 eV and $L_2$ ($e_g$) at 463.4 eV, with least O–K edge intensity at 531 eV, which corresponds to the pure Ti$^{3+}$ oxidation state. Moving down toward Ti/$a$-STO interface, Ti–$L_{2,3}$ changes to slightly a higher value and peak broadening is observed. Moreover, less pronounced peak splitting and minor O–K peak is clearly observed. This can be attributed to Ti$^{3+}$ valence. It should be noted that Ti$^{5+}$ and Ti$^{2+}$ peaks are similar and occur in close proximity; therefore, at the interface Ti$^{2+}$ valence might also be present. In the bulk $a$-STO layer, peak splitting is observed for both $L_1$ and $L_2$ peaks appearing as four peaks: $L_1$ ($t_{2g}$) at 458.6 eV, $L_1$ ($e_g$) at 460.35 eV, $L_2$ ($t_{2g}$) at 464.14 eV, and $L_2$ ($e_g$) at 465.8 eV. This affirms the Ti$^{4+}$ oxidation state, confirming the stoichiometric nature of STO film over the bulk of $a$-STO layer.[29] This proves that the Ti oxidation state changes from Ti$^{3+}$ in metal TE, to Ti$^{4+}$ at the Ti/$a$-STO interface, and then to Ti$^{4+}$ in the bulk of $a$-STO. It further affirms that the Ti layer near the interface oxidizes due to the electrochemical redox reaction, forming TiO$_x$ (Ti$_2$O$_3$-like) interfacial layer.[29,32]

Toward the $a$-STO/$a$-VO$_x$ interface, we clearly observe four peaks of Ti corresponding to Ti$^{4+}$ (or stoichiometric $a$-STO). At the same location we also observe vanadium with $L_1$ peak at 525.5 eV and $L_2$ peak at 519.2 eV. Here, the O–K edge intensity is only slightly higher compared with the bulk $a$-STO layer. This shows the diffusion of vanadium ions into $a$-STO layer. Further down at the interface, we observe two Ti–$L_{2,3}$ ($e_g$) peaks corresponding to the Ti$^{3+}$ valence. This affirms the substoichiometric nature (oxygen deficient) of $a$-STO at the $a$-STO/$a$-VO$_x$ interface. $V–L_{2,3}$ peak positions at this location are same as before. Moving toward the bulk, $a$-VO$_x$ peak position shifts to a slightly higher value with narrower peaks compared with those at the interface. It should be noted that the oxidation phases of vanadium from $V^{2+}$ to $V^{5+}$ occur in a short range of 1.3 eV with slightly different positions centered with $V–L_3$ at 525 eV and $V–L_2$ at 518 eV.[44]

Different oxidation phases of vanadium oxide are distinguished by observing the O–K peak edge.[14] Due to the experimental limitation, achieved O–K peaks are noisy and it is not possible to resolve the exact oxidation phase of vanadium oxide. However, it should be noted that the $V–L_{2,3}$ and O–K peak position at the interface of $a$-STO/$a$-VO$_x$ and the bulk of $a$-VO$_x$ is nearly constant. Further, XPS analysis of our as-deposited $a$-VO$_x$ and $a$-STO is presented in Figure S1, Supporting Information. The composition profile graph of EELS analysis presented in Figure S2, Supporting Information, suggests that the thickness of $a$-STO and $a$-VO$_x$ interface is $\approx$ 14 nm.

To summarize the EELS analysis, we distinguished the cross-point device stacks in seven regions as Ti$^0$ pure metal phase in TE, Ti$^{4+}$ (Ti$_2$O$_3$-like) phase at the Ti/$a$-STO interface, bulk $a$-STO, vanadium-diffused $a$-STO near $a$-STO/$a$-VO$_x$ interface, substoichiometric $a$-STO and $a$-VO$_x$ at $a$-STO/$a$-VO$_x$ interface, bulk $a$-VO$_x$, and pure Pt metal phase. Pt peak being out of the dispersion window does not show any peak in Figure 5a.

2.4. Switching Mechanism

Finally, considering all the electrical characterizations and the compositional analyses, we provide insights into the possible switching mechanism of the hybrid device. Nonvolatile BRS in $a$-STO is associated with the filamentary breakdown and exchange of oxygen ions at the top electrode (Ti) and oxide interface, which has been reported for four distinctive regions of elemental distribution along the device stacks—Ti$^0$, Ti$_2$O$_3$, $a$-STO, and Pt.[29,34] Our EELS data also show similar elemental distribution and evolution of Ti phases along the Ti/$a$-STO interface. Moreover, apolar TS in $a$-VO$_x$-based devices has been reported due to the combination of filamentary breakdown and
insulator-to-metal transition in local crystal islands of c-VO₂ formed within or near the filament.\textsuperscript{[23]} As proved in the aforementioned sections, the hybrid device retains electrical properties of both the individual components. It has nonvolatile nature like a-STO along with the TS behavior like a-VOₓ.

These similarities lead us to propose a switching mechanism of the hybrid device as a combination of the individual switching mechanisms of a-STO and a-VOₓ, shown in Figure 6. In the hybrid device, electroforming would be associated with the filamentary break down, as shown in Figure 1b. In a-STO region, the filament of oxygen vacancies and metallic ion would be present,\textsuperscript{[38]} whereas it would be made up of vanadium and oxygen ions in the a-VOₓ region.\textsuperscript{[23]} Local crystal islands of c-VO₂ would form within or near the filament region of a-VOₓ as a part of the electroforming process, as shown in Figure 6b. After electroforming, c-VO₂ crystal islands in a-VOₓ would undergo the insulator-to-metal transition, resulting in threshold behavior. When negative bias is applied to BE, c-VO₂ would transit in the metallic state, as shown in Figure 6d. At this stage, the maximum potential drop would occur across a-STO as the resistance of a-VOₓ region would be very low due to metallic c-VO₂. This would provide sufficient energy for oxygen ions to migrate across the Ti and a-STO interface, resulting in rupturing of the filament permanently, which resets the device to high resistance state (HRS), as shown Figure 6e. When the electrical bias is removed, c-VO₂ returns back to the insulating state due to its volatile nature, as shown in Figure 6f. For the reverse polarity cycle, first, c-VO₂ crystal islands transit into the metallic state; second, filament is formed, which sets the device to LRS; and finally, c-VO₂ reverts back into the insulating state when the bias is removed. Polarity of the bias does not affect the insulator-to-metal transition in c-VO₂ as the TS in a-VOₓ is volatile, apolar, and symmetric.\textsuperscript{[23]} However, it affects the direction of oxygen-ion migration at the interface of Ti/a-STO, resulting in the nonvolatile nature of the hybrid device.

This switching mechanism for the hybrid device is proposed by analyzing the achieved results and comparing it with the reported studies. A simple elemental analysis is presented at the each step of switching mechanism in Figure 6. The claim of filamentary switching is supported by the area dependence of switching voltages, as shown in Figure 4b. As the switching voltages of devices with area in a large range from 200 to 90 mm\textsuperscript{2} are within a small range from ±2 to ±4 V, it strongly supports filamentary switching in the hybrid device.\textsuperscript{[46]} However, the

![Figure 6. Schematic representation of the proposed switching mechanism for hybrid device. In the hybrid multilayer device, in (a), a conductive filament based on oxygen and metal ions forms due to electroforming, as shown in (b). As a part of electroforming, localized c-VO₂ forms within the filament region of a-VOₓ, in (c). Localized c-VO₂ undergoes the insulator-to-metal transition (d), resulting in threshold behavior of the hybrid device. When c-VO₂ is in metallic state, maximum potential drop occurs across a-STO region. This provides enough energy for oxygen ions to migrate across the Ti and a-STO interface, resulting in rupturing of the filament (e) (or formation of the filament (d)), and remain in that state when bias is removed, and c-VO₂ returns back to insulator state (f) (for formed filament) and thereby, a nonvolatile memory nature of the hybrid device. A simple elemental analysis is presented at each step of electroforming and switching. A color scheme is used to highlight the resistance state at each step of the switching cycle. Here, Rf_STO represents the filament resistance in a-STO region and Rf_VO represents the filament resistance in a-VOₓ region which including localized c-VO₂ islands.](image-url)
proposed mechanism assumes that a-STO/a-VO₃ interface plays insignificant role in the overall switching mechanism and in-depth investigations are required in the future to ascertain switching mechanism propensity.

3. Conclusion

We present a hybrid cross-point device structure with functional oxide as a bilayer of a-VO₃ and a-STO. This device retains the TS of a-VO₃ and nonvolatile BRS of a-STO, showing one-selector-one-resistor switch equivalent characteristics. The hybrid devices show a mean switching ratio of 8, nonlinearity factor of 4.8, stability for 6000 endurance cycles, RM of 47.8%, and a potential array size of $10^7 \times 10^4$ when 10% RM is considered. The quantitative analysis of selector effect in the hybrid device is calculated using a novel approach of comparing the nonzero-crossing characteristics of the hybrid, a-STO and a-VO₃ devices. This shows significant six times reduction in the OFF current, whereas the nonlinearity factor remains nearly constant—two aspects deciding the overall sneak-path current. Conventionally, this suggests minimal improvement in the readout margin and array size of hybrid device; however, we believe that the direct effect of OFF current on the array size has to be investigated in future.

Further, EELS analysis is presented over the cross section of the device to understand elemental distribution along the cross section of the device and at interfaces. The results present the seven distinctive regions of elemental distribution across the stacks of the hybrid device. Further, we suggest the possible switching mechanism along with simple elemental analysis. This work highlights the ideal selector properties of vanadium oxide (a-VO₃), along with one of the leading resistive switching materials a-STO in a cross-point hybrid architecture for simple fabrication and biasing scheme, unlike the conventional 1S1R structure. It further provides the elemental distribution across the interface of two distinctive metal oxides a-STO and a-VO₃, which can be useful for diverse electronic applications.

4. Experimental Section

**a-VO₃ Thin-Film Deposition:** Thin films ($\approx 100$ nm) of a-VO₃ were deposited on Pt/Ti (15/5 nm) using pulsed direct current (DC) sputtering. Primary target used was vanadium (V), with a substrate of thermally grown silicon dioxide (300 nm) on silicon (SiO₂/Si). Deposition was conducted in 30% oxygen atmosphere at room temperature with a sputtering pressure of 4 mTorr (from the base pressure of $<10^{-7}$ Torr). More details about deposition conditions and film quality can be found in related literature.\(^{[47]}\)

**a-STO Thin-Film Deposition:** Thin films of 100 nm a-STO were deposited on Pt/Ti/SiO₂ (15:5:300 nm)-coated Si substrates using radio frequency (RF) magnetron sputtering at room temperature, from a stoichiometric ceramic target. Thin films were sputtered in pure argon atmosphere with a sputtering pressure of 5 mTorr, from a base pressure of $<10^{-7}$ Torr.

**Cross-Point Device Fabrication:** Fabrication of cross-point devices was conducted using three-step photolithography and lift-off process. In the first step, prepatterned BE was deposited with 15 nm Pt with 5 nm Ti adhesion in Lesker PVDTS electron beam evaporator system. Subsequently, functional oxide thin films were sputtered and lift-off was conducted. Finally, Ti/Pt (20:200 nm) TE was deposited on prepatterned TE and lifted off.

**Electrical Characterization:** Electrical characterization of cross-point devices was conducted using Keithley 4200 source meter with two-probe DC I–V measurements. All characterizations were conducted with BE biased and opposite electrode (TE) connected to system ground.

**TEM and EELS:** Device cross-section TEM imaging and EELS analysis were conducted using FEOL 2100 with an attached Tridium Gatan image filter. For cross-sectional TEM analysis, lamella was prepared by focused-ion beam (FIB) cuts using FEI Scios DualBeam system from the fabricated pristine devices.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest**

The authors declare no conflict of interest.

**Data Availability Statement**

The data that supports the findings of this study are available in the supplementary material of this article.

**Keywords**

hybrid bilayer devices, resistive memories, strontium titanate, vanadium oxide

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