Understanding modes of negative differential resistance in amorphous and polycrystalline vanadium oxides

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Abstract

Metal-oxide-metal devices based on amorphous VOₓ are shown to exhibit one of two distinct negative differential resistance (NDR) characteristics depending on the maximum current employed for electroforming. For low compliance currents they exhibit a smooth S-type characteristic and have a temperature-dependent device resistance characterised by an activation energy of 0.25 eV, consistent with conduction in polycrystalline VO₂, while for high-compliance currents they exhibit an abrupt snap-back characteristic and a resistance characterised by an activation energy of 0.025 eV, consistent with conduction in oxygen deficient VOₓ. In both cases, the temperature dependence of the switching voltage implies that the conductivity change is due to the insulator-metal transition in VO₂. From this analysis it is concluded that electroforming at low currents creates a conductive filament comprised largely of polycrystalline VO₂, while electroforming at high currents creates a composite structure comprised of VO₂ and a conductive halo of oxygen deficient VOₓ. The effect of electroforming on the NDR mode is then explained with reference to a lumped element model of filamentary conduction that includes the effect of a parallel resistance created by the halo. These results provide new insight into the NDR response of vanadium-oxide-based devices and a basis for designing devices with specific characteristics.

1. Introduction

Current controlled negative differential resistance (NDR) in metal-oxide-metal devices is of interest as the basis of nanoscale relaxation oscillators for use as solid-state neurons in neuromorphic computing arrays.¹,² The as-fabricated devices are generally in a high resistance state and require a one-off electroforming step to initiate the NDR response.³,⁴ This is typically

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achieved by subjecting the film to a voltage or current stress sufficient to form a filamentary conduction path through the film (i.e. soft dielectric breakdown), a process mediated by the generation, drift and diffusion of atoms and ions in response to the applied electric field and local Joule heat.\textsuperscript{5, 6} The size, resistance and stability of the resulting filaments depend critically on the forming conditions, and particularly on the maximum forming current and the associated temperature rise caused by Joule heating.\textsuperscript{7, 8} The high temperatures associated with electroforming can also cause crystallisation of amorphous films and compositional or structural changes at the oxide/electrode interface that affect the final state of the electroformed device and its switching characteristics.\textsuperscript{9, 10} As a consequence, understanding details of the electroforming process is an essential requirement for developing devices with specific characteristics.

In amorphous vanadium-oxide based devices the NDR response is generally attributed to the insulator-metal transition in VO\textsubscript{2} on the assumption that this phase is crystallised within the filamentary conduction path during electroforming.\textsuperscript{9} Such devices generally exhibit smooth S-type NDR due to the heterogeneous nature of the IMT transition and the evolution of the temperature distribution during current-controlled testing.\textsuperscript{11} However, they can also exhibit an abrupt snap-back characteristic under certain condition, similar to that observed in NbO\textsubscript{x} based devices.\textsuperscript{7, 12} This novel NDR mode has the potential to offer new device functionality but its origin continues to be debated.\textsuperscript{12, 13}

In this study, we show that electroforming can be used to control the NDR characteristics of amorphous VO\textsubscript{x} films and that the snap-back response can be understood from the filament microstructure and its impact on the effective circuit of the device.

2. **Experimental Details**

Two device structures were employed for these studies: metal-oxide-metal (MOM) capacitor structures fabricated with a common bottom electrode (BE) and top electrodes (TE) of 100\(\mu\)m diameter circles defined by a shadow mask; and cross-point devices (2\(\mu\)m x 2\(\mu\)m and 5\(\mu\)m x 5\(\mu\)m) fabricated using step-by-step photolithography, as shown in Figure 1.\textsuperscript{14} In both cases, the devices were fabricated on thermally oxidised (100nm SiO\textsubscript{2}) Si wafers by sequential layer deposition. The bottom electrodes consisted of a 10 nm-thick Ti adhesion layer and a 50 nm-thick Pt contact layer deposited by sequential e-beam evaporation. A 70 nm thick functional oxide layer of either amorphous VO\textsubscript{x} or polycrystalline VO\textsubscript{2} was then deposited by reactive
sputter deposition from a V target using an O₂/Ar ambient maintained at a pressure of 2.3 (or 1.5) mTorr using Ar/O₂ flow rates of 58/2 (or 58/10) sccm. Amorphous VOₓ films (a-VOₓ) were achieved by maintaining the substrates at room temperature and polycrystalline VO₂ films (pc-VO₂) were achieved by post-annealing the film at 450 °C in a partial vacuum (1.5 Torr air). The devices were completed by adding top electrodes consisting of a 5 nm-thick Ti layer and a 25 nm Pt layer.

The as-deposited oxide films were characterised by grazing incident angle X-ray diffraction (GI-XRD), atomic force microscopy (AFM), Raman spectroscopy (RS) and electron Rutherford backscattering spectrometry (eRBS). Electrical measurements were performed with an Agilent B1500A semiconductor parameter analyser attached to a Signatone probe station (S-1160) and were undertaken in air by applying voltages to top electrode while grounding the bottom electrode.

Figure 1: (a) Schematic showing the test structure of Pt/Ti/a-VOₓ/Pt devices and (b) scanning electron microscopy image of Pt/Ti/pc-VO₂/Pt cross-point device showing four 20 µm x 20 µm devices with a common bottom electrode with 3D schematic of the cross-point area.

3. Experimental Results and Discussion

3.1 Composition and Structure of Films

Figure 2 shows GI-XRD spectra and AFM images of the as-deposited a-VOₓ and pc-VO₂ films, together with Raman spectra from the pc-VO₂ film as a function of temperature. The GI-XRD spectrum from the pc-VO₂ film has peaks corresponding to (011), (220), and (022) planes of monoclinic VO₂, while that from a-VOₓ film is essentially featureless, consistent with the film being amorphous. (The only diffraction peaks observed in this case are from the underlying Pt substrate). Temperature dependent Raman analysis showed that the pc-VO₂ film underwent a thermally induced phase transition at temperatures between 40°C and 80°C, consistent with the well-known insulator-metal phase transition in VO₂. The surface morphology and roughness of
the films was determined from AFM images and was similar for both films, with the RMS roughness measured to be 2.1 and 2.3 nm for the a-VOₓ and pc-VO₂ films, respectively. These results, combined with eRBS analysis show that the pc-VO₂ films are polycrystalline and composed of monoclinic VO₂, while the VOₓ films are amorphous and have a composition close to V₂O₅ (i.e. x~2.5).

3.2 Electrical characterization

As-fabricated devices were highly resistive, with resistances of order several MΩ, and required a one-off electroforming step to initiate threshold switching, as shown in Figure 3. This was achieved by scanning either the voltage or the current and detecting the abrupt conductivity

Figure 2: (a) GIXRD of pc-VO₂ and a-VOₓ films deposited on Pt, (b) Raman spectra from pc-VO₂ as a function of temperature, and (c) AFM images of the pc-VO₂ and a-VOₓ films.
change indicative of filament formation, and typically reduced the device resistance by around an order of magnitude, consistent with the creation of a filamentary conduction path in the oxide layer. For voltage controlled electroforming the maximum current ($I_{CC}$) was limited to avoid device damage. Immediately following electroforming the devices exhibited symmetric threshold switching under voltage controlled testing, with threshold voltages in the range from $\pm0.45$ V to $\pm2.2$ V. Similar behavior has previously been reported in both lateral and vertical device structures and is generally attributed to the thermally induced IMT in VO$_2$ and the associated positive feedback created by Joule heating.$^{16-18}$ In the case of a-VO$_x$ devices, this is predicated on the assumption that the VO$_2$ phase is crystallized within the amorphous film during electroforming$^9$.

Figure 3: Electroforming (dashed line) and subsequent threshold switching characteristics (solid line) for (a) an a-VO$_x$ capacitor structure (100 $\mu$m diameter) and (b) a pc-VO$_2$ cross-point device (2$\mu$m x 2 $\mu$m).
Figure 4a shows corresponding current-controlled I-V characteristics for these devices. In this case, the current is constrained by the measurement system so that the increase in conductivity is self-limiting and the I-V characteristics vary continuously and the regions of NDR reflect the fact that the conductivity increases superlinearly with current (temperature). Given the significant difference in the initial film properties and device structures, the characteristics of the pc-VO₂ and a-VOₓ devices are remarkably similar, reflecting the filamentary nature of the conduction process and the common origin of the conductivity change.

![Figure 4: (a) S-type NDR in an a-VOₓ capacitor structure and inset showing similar behaviour in a 2µm × 2µm pc-VO₂ cross-point device. (b) Snap-back NDR characteristics an a-VOₓ capacitor structure and inset showing similar behaviour in a 5µm × 5µm pc-VO₂ cross-point device.](image)

Of particular interest in this study is the fact that these devices can also exhibit a discontinuous ‘snap-back’ characteristic, such as that shown in Figure 4b. This is an alternative switching
mode characterised by an abrupt increase in conductivity as the current reaches its threshold value and an abrupt reduction in conductivity as the current returns to its hold value. For the cases shown in Figure 4b, this hysteretic snap-back mode was effected by increasing the device area of the pc-VO2 cross-point device from 4 µm$^2$ to 25 µm$^2$, and by increasing the electroforming compliance current for the a-VOx capacitor structure. In both cases, the transition is associated with a reduction in the device resistance but this alone does not explain the origin of snap-back response.

### 3.3 Temperature dependence

To gain further insight into these switching modes I-V characteristics were also investigated as a function of temperature, and Figure 5 shows typical results. The subthreshold I-V characteristics of both pc-VO2 and a-VOx devices are well modelled by a trap-limited conduction model (e.g. Poole-Frenkel conduction$^{20}$), while voltage controlled threshold switching and current-controlled snap-back characteristics of a-VOx devices serve to illustrate the systematic reduction of the threshold voltages ($V_{th}$) and hold voltages ($V_h$) with increasing temperature. The inset in Figure 5b also highlights the presence of discrete resistance changes during the metal to insulator transition, as previously reported for both thermal and voltage cycling of VO2 devices where it was attributed to the heterogeneous nature of the transition$^{11, 21}$.

![Figure 5](image)

**Figure 5:** (a) Sub-threshold I-V characteristics of a post-formed a-VOx capacitor structure as a function of temperature, and inset showing similar behaviour for a 5µ × 5µ pc-VO2 cross-point device; (b) voltage-controlled threshold switching response of an a-VOx device as a function of temperature; and (c) Current-controlled NDR response of an a-VOx device as a function of temperature.

Results from such measurements are summarized in Figure 6 which shows the temperature dependence of $V_{th}$ and an Arrhenius plot of the sub-threshold device resistance for selected pc-
VO$_2$ and a-VO$_x$ devices. For switching based on a thermally induced metal-insulator transition, $V_{th}$ is expected to decrease with increasing device temperature ($T_0$) due to the fact that the transition temperature ($T_{IMT}$) can be achieved at lower power$^{22, 23}$. In this case the filament temperature ($T$) can be approximated by a lumped element model such that $T = T_0 + R_{therm}IV$, where $R_{therm}$ is the thermal resistance of the filament, $I$ is the device current and $V$ is the device voltage. Using Ohm’s law and assuming a thermally activated device resistance of the form $R = R_0e^{-E_a/kT}$ then reveals that $V_{th}^2 = \frac{R_0}{R_{therm}}(T_{IMT} - T_0)e^{E_a/kT_{IMT}}$. i.e. $V_{th}^2$ scales linearly with the device temperature $T_0$ and goes to zero as $T_0$ approaches $T_{IMT}$. As shown in Figure 6a, the measured $V_{th}$ for pc-VO$_2$ and a-VO$_x$ devices satisfies this equation and has an intercept in the range 340-350 K, consistent with the IMT in VO$_2$.$^{21, 24}$ Significantly, this temperature is similar for pc-VO$_2$ and a-VO$_x$ devices that exhibit continuous S-type NDR and for the a-VO$_x$ device that exhibits abrupt snap-back characteristics. This confirms the role of VO$_2$ in the switching of a-VO$_x$ devices and suggests that both the S-type and snap-back characteristics have a common origin.

![Graph](image)

**Figure 6a**
- $V_{th}^2$ vs. $T$ for different types of devices.
- pc-VO$_2$, a-VO$_x$ (S-type), a-VO$_x$ (snap-back).

**Figure 6b**
- Log of $R$ vs. $1/kT$ for different $E_a$ values.
- pc-VO$_2$, a-VO$_x$ (S-NDR), a-VO$_x$ (snap-back), $E_a = 0.25$ eV, $E_a = 0.025$ eV.
Figure 6: (a) Temperature dependence of the threshold voltage, and (b) an Arrhenius plot of the sub-threshold resistance for a-VOₓ devices that exhibit S-type and snap-back NDR and for a 5µm x 5µm pc-VO₂ device that exhibits snap-back NDR.

The associated Arrhenius plot shows that the change in sub-threshold resistance of the devices that exhibit S-type NDR is well characterised by a single activation energy of ~0.25 eV, consistent with previously reported values for conduction in polycrystalline VO₂. This further reinforces the view that VO₂ is the dominant phase both the a-VOₓ and pc-VO₂ devices. However, the sub-threshold resistance of the a-VOₓ device that exhibits a snap-response is characterised by an activation energy of ~0.025 eV, similar to that observed for the low resistance state of oxide-based resistive switching devices following an electroforming or a ‘set’ operation. This is a particularly revealing as it suggests that subthreshold conduction is dominated by a high conductivity path through the oxide film even though the device exhibits a switching response characteristic of the IMT in VO₂.

3.4 Proposed model

Both pc-VO₂ and a-VOₓ devices were shown to exhibit two distinct modes of CC-NDR, a smooth S-type mode or an abrupt snap-back mode, with the dominant switching mode dependence on the device area and the electroforming conditions. To understand this behaviour we draw on results from parallel studies in NbOₓ-based devices where the NDR is attributed to the temperature dependence of trap-assisted conduction (e.g. Poole-Frenkel conduction).

Using NbOₓ-devices as a prototypic example of filamentary threshold switching we showed that the snap-back mode of CC-NDR can arise from a current redistribution process in which the current flowing in the region surrounding the conductive filament abruptly concentrates within the filament as it exhibits NDR in response to local Joule heating. This can be understood by representing the device by a core-shell structure in which the core represents the high conductivity filament and the shell represents the parallel resistance due to conduction in the surrounding film. Simple circuit analysis then shows that current redistribution is controlled by the relative magnitudes of the NDR of the core, R_NDR, and the resistance of the shell, R_S, with continuous S-type characteristics observed for R_S>R_NDR and abrupt snap-back characteristics observed for R_S<R_NDR. This implies that the current in the surrounding area must be comparable to that in the filament in order to observe the snap back response and highlights the fact that the dominant behaviour will depend on the resistivity, thickness and...
area of the oxide film. Significantly, this analysis is independent of the process responsible for NDR.

In the present case, pc-VO$_2$ films were found to be much more conductive than a-VO$_x$ films and, as a consequence, the device resistance of 2 $\mu$m x 2 $\mu$m pc-VO$_2$ cross-point devices was comparable to that of 100 $\mu$m diameter a-VO$_x$ capacitor structures. In both cases these devices exhibited continuous S-type NDR when electroformed using low compliance currents. However, when the area of the pc-VO$_2$ cross-point devices was increased to 5 $\mu$m x 5 $\mu$m (a factor of 6.25) they exhibited a snap-back response, and when the 100 $\mu$m diameter a-VO$_x$ capacitor structures were electroformed using high compliance currents they also exhibited a snap-back response. Within the framework of the core-shell model the behaviour of the pc-VO$_2$ cross-point devices can be understood by accounting for the effect of the device area on the magnitude of the shell resistance, $R_s$. i.e. For the small area devices $R_s>R_{\text{NDR}}$ and the devices exhibit continuous S-type characteristics, while for the large area devices $R_s<R_{\text{NDR}}$, and they exhibit snap-back characteristics. Indeed, similar behaviour has previously been reported for NbO$_x$ devices.$^5,7,12$

The effect of electroforming on the a-VO$_x$ devices requires further explanation. In this case, the device area was fixed and it is tempting to attribute the snap-back response to a change in $R_{\text{NDR}}$. However, we have found no clear correlation between $R_{\text{NDR}}$ and the electroforming conditions. Instead, we refer to the temperature dependent measurements in Figure 6 which show that sub-threshold conduction in a-VO$_x$ devices electroformed using high compliance currents is characterised by an activation energy of 0.025 eV, much lower than the 0.25 eV observed for devices formed at low compliance currents and for comparable pc-VO$_2$ devices. Despite this low activation energy, the NDR response of the devices remains consistent with the IMT in VO$_2$, suggesting that the filament consists of both highly conductive VO$_x$ and pc-VO$_2$. Given the nature of the electroforming process, the filament is expected to have radial symmetry and to consist of a central pc-VO$_2$ core and a halo of substoichiometric VO$_{x-\delta}$.10,29

The effect of electroforming can then be understood on the basis that the relative sizes of the pc-VO$_2$ core and VO$_{x-\delta}$ halo depend on the compliance current, with low compliance currents producing filaments that are dominated by pc-VO$_2$ and high compliance currents producing filaments with pc-VO$_2$ core and a significant VO$_{x-\delta}$ halo.
Figure 7 shows schematic representations of the proposed filament structures in a-VOₓ devices electroformed with ‘low’ and ‘high’ compliance currents, together with an equivalent electrical circuit. The relative diameters of the core and halo regions are assumed to increase with increasing forming current, consistent with the observed reduction in filament resistance and its temperature dependence. From an electrical perspective, the device can then be considered as three parallel resistors: one associated with the core and having a temperature dependent resistance governed by the heterogeneous IMT of VO₂; the second with the halo region and having a resistance determined by the electroforming conditions; and the third with the surrounding film and having a resistance determined by the stoichiometry, thickness and area of the oxide film. To a reasonable approximation the resistance of the halo and the surrounding film can be treated as constants so that the model reduces to the core-shell model discussed earlier, with the core represented by a temperature dependent resistor and the shell by a fixed parallel resistor of magnitude: \( R_s = \frac{R_h R_f}{R_h + R_f} \), where \( R_h \) is the effective resistance of the filament halo and \( R_f \) the effective resistance of the surrounding film. Based on this model, the observation that the a-VOₓ devices exhibit S-type NDR at low compliance currents and snap-back NDR at high compliance currents can also be attributed to a change in \( R_s \), albeit from change in the halo resistance as a result of electroforming rather than a change in device area.

\[
R_s = \frac{R_h R_f}{R_h + R_f}
\]

4. **Summary and Conclusions**

The voltage controlled threshold switching and current-controlled NDR behaviour of electroformed a-VOₓ and pc-VO₂ devices was investigated. Temperature dependent electrical
measurements were consistent with the IMT in VO$_2$, and with the crystallisation of this phase within the a-VO$_x$ films during electroforming. Following electroforming each device type exhibited two distinct modes of NDR, a continuous S-type response and an abrupt snap-back response, depending on the device area and electroforming conditions. This behaviour was interpreted with reference to a simple core shell model of filamentary conduction in which the core represented the high-conductivity filament and the shell represented a parallel resistance due to the surrounding film. This predicted a transition between the S-type and snap-back behaviour based on the relative magnitudes of core NDR the shell resistance. For the pc-VO$_2$ devices the transition was demonstrated by using the device area to vary the shell resistance, as previously reported for a-NbO$_x$ devices. However, in the case of a-VO$_x$ devices it was controlled by the electroforming conditions. In that case, electroforming with high compliance currents was shown to produce composite filaments that were characterised by a high conductivity and weak temperature dependence at sub-threshold current but by the IMT of VO$_2$ at high currents. This was represented by a pc-VO$_2$ core and a surrounding substoichiometric halo, with the high conductivity halo acting to reduce the overall shell resistance and thereby control the transition between S-type and snap-back modes. Significantly, these results show that the snap-back characteristic is a generic response of systems that exhibit NDR

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6. Data Availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.
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