A numerical analysis and experimental demonstration of a low degradation conductive bridge resistive memory device

Berco, Dan; Chand, Umesh; Fariborzi, Hossein

2017

Berco, D., Chand, U., & Fariborzi, H. (2017). A numerical analysis and experimental demonstration of a low degradation conductive bridge resistive memory device. *Journal of Applied Physics*, 122(16), 164502-.

<https://hdl.handle.net/10356/86697>

<https://doi.org/10.1063/1.5008727>

© 2017 American Institute of Physics (AIP). This paper was published in *Journal of Applied Physics* and is made available as an electronic reprint (preprint) with permission of American Institute of Physics (AIP). The published version is available at: [<http://dx.doi.org/10.1063/1.5008727>]. One print or electronic copy may be made for personal use only. Systematic or multiple reproduction, distribution to multiple locations via electronic or other means, duplication of any material in this paper for a fee or for commercial purposes, or modification of the content of the paper is prohibited and is subject to penalties under law.

Downloaded on 26 Apr 2023 21:48:26 SGT

A numerical analysis and experimental demonstration of a low degradation conductive bridge resistive memory device

Dan Berco,^{1,a)} Umesh Chand,² and Hossein Fariborzi²

¹*Nanyang Technological University, School of Electrical and Electronic Engineering, 50 Nanyang Avenue, Singapore, Singapore 639798*

²*Computer, Electrical and Mathematical Sciences & Engineering (CEMSE) Division, King Abdullah University of Science and Technology (KAUST), Thuwal, Saudi Arabia*

(Received 24 March 2017; accepted 8 October 2017; published online 23 October 2017)

This study investigates a low degradation metal-ion conductive bridge RAM (CBRAM) structure. The structure is based on placing a diffusion blocking layer (DBL) between the device's top electrode (TE) and the resistive switching layer (RSL), unlike conventional CBRAMs, where the TE serves as a supply reservoir for metallic species diffusing into the RSL to form a conductive filament (CF) and is kept in direct contact with the RSL. The properties of a conventional CBRAM structure (Cu/HfO₂/TiN), having a Cu TE, 10 nm HfO₂ RSL, and a TiN bottom electrode, are compared with a 2 nm TaN DBL incorporating structure (Cu/TaN/HfO₂/TiN) for 10³ programming and erase simulation cycles. The low and high resistive state values for each cycle are calculated and the analysis reveals that adding the DBL yields lower degradation. In addition, the 2D distribution plots of oxygen vacancies, O ions, and Cu species within the RSL indicate that oxidation occurring in the DBL-RSL interface results in the formation of a sub-stoichiometric tantalum oxynitride with higher blocking capabilities that suppresses further Cu insertion beyond an initial CF formation phase, as well as CF lateral widening during cycling. The higher endurance of the structure with DBL may thus be attributed to the relatively low amount of Cu migrating into the RSL during the initial CF formation. Furthermore, this isomorphic CF displays similar cycling behavior to neural ionic channels. The results of numerical analysis show a good match to experimental measurements of similar device structures as well. *Published by AIP Publishing.* <https://doi.org/10.1063/1.5008727>

INTRODUCTION

Resistive non-volatile RAM (RRAM) technology has evolved in the past decade to allow extremely compact, low power devices,^{1–3} which are suitable candidates for implementation of a synthetic synapse that serves as a basic building block for artificial neural networks. In addition, their compatibility with CMOS technology makes them appealing devices for the implementation of high bit densities brain-inspired computing systems.^{4–6}

The conduction mechanism of oxygen ion-based RRAM is due to the formation and rupture of a percolation like conduction path,⁷ commonly referred to as a conductive filament (CF), while that of a conductive bridge RAM (CBRAM) is obtained through the manipulation of nano-scale quantities of metal in thin dielectric films or solid electrolytes.⁸ A CF that consists of trap like oxygen vacancies (marked OV with a spatial density N_{OV}) and mobile oxygen species (marked O with a spatial density N_O)^{9,10} forms a conduction path through the dielectric film. Alternatively, the metallic species based conduction mechanism of the metal-ion based CBRAM^{11,12} (marked Cu with a spatial density N_{Cu}) make it a favorable candidate for future non-volatile memory (NVM) devices due to fast switching, high endurance, and scalability. The operation of CBRAM device is believed to be based on the formation and rupture of a CF consisting of the oxidation and the

reduction of metal atoms in the resistive switching layer (RSL) in addition to the OV, where metal oxides are used as RSL.

The understanding of the physical mechanisms that determine device reliability along with the enhancement of endurance^{13,14} plays a key role in the successful implementation of artificial synapses by RRAMs. This work presents a numerical analysis of a novel CBRAM structure designed to overcome the degradation mechanisms associated with conventional CBRAM due to long run operation and cycling. The structure is based on a counter intuitive approach of placing a diffusion blocking layer (DBL) between the device's top electrode (TE) and the RSL, unlike conventional CBRAMs where the TE serves as a supply reservoir for metallic species diffusing into the RSL to form a CF and is kept in direct contact with the RSL. The theoretical framework and modeling considerations of the analysis method are first presented. The properties of a conventional CBRAM are then compared with a DBL incorporating structure for 10³ programming and erase simulation cycles. The simulation results are further compared with the experimental measurements of similar device structures.

MODELING

A conventional CBRAM structure (Cu/HfO₂/TiN) was chosen as a benchmark platform to demonstrate the performance improvement resulting from adding a DBL to a metal-insulator-metal device. The algorithm used in this work is detailed in the previous publications.^{9,11} The key aspects of

^aAuthor to whom correspondence should be addressed: danny.barkan@gmail.com

the model in reference to the obtained results are thus only briefly discussed herein. For further information, the reader is advised to consult those publications. In this work, the properties of a 2 nm TaN DBL incorporating structure (Cu/TaN/HfO₂/TiN marked: ST1), having a Cu TE, 10 nm thick HfO₂ RSL and a TiN bottom electrode (BE), are compared with a similar conventional CBRAM structure (Cu/HfO₂/TiN marked: ST2) for 10³ cycles.

The approach is demonstrated on monoclinic hafnium oxide (HfO₂) based devices and simulate both the forming and reset operation modes, where a CF is formed and ruptured respectively. The RSL is modeled using a 2D rectangular structure, having width W and height H , while divided into a uniform grid (of $\delta = 0.5$ nm spacing) with cyclic boundary conditions in the x direction. Each grid site $n \in N$ is represented by its spatial coordinates $n = (x, y)$ on which the local potential φ_n , temperature T_n , $N_{o,n}$, $N_{ov,n}$ and electric conductivity σ_n are calculated. The dimensions of the RSL are chosen as $W = 20$ nm and $H = 10$ nm. The top and bottom control electrodes (TE and BE) are located at $y = H$ and $y = 0$, respectively. A constant voltage is applied to the TE, corresponding to either programming or erase operational modes, while the bottom one is grounded.

The BE material (TiN) is considered as inert and the TE material (Cu) acts as the source of metallic species migrating into the RSL. The simulation is done while manipulating the density functions of O, OV, and diffusing Cu species, starting from an initial random state. The values of the parameters used in the simulation as well as the modeling considerations are elaborated in the previous publications^{9,11} and are listed in Table I.

The interaction of the TaN layer with O species in ST1 is calculated as a function of $T_{(x,H)}$ and $N_{o,(x,H)}$ and modeled by an independent data structure indicating the interfacial concentration in the DBL (marked: $N_{o,TaN}$). The determination of Cu diffusion coefficients from the TE/TaN and into the RSL, and $N_{Cu,(x,H)}$, is thus made as a function of this interfacial local oxidation.

FORMULATION

This section highlights the extension made to the algorithm detailed in Ref. 9 following the specific requirements

TABLE I. Parameter values used in this work.^{9,11,17}

Symbol	Description	Value
f_o	O, Cu ion vibration frequency	10 ¹³ Hz
Z	O ion charge number	2
E_{ac}	Conductance activation energy	50 meV
D_o	O ion diffusivity factor	2 m cm ² /s
E_o	Cu oxidation activation energy	3.1 eV
E_r	Cu reduction activation energy	3.1 eV
E_{chf}	Cu-HfO ₂ insertion activation energy	1.3 eV
E_{cov}	Cu-OV insertion activation energy	~0 eV
E_h	O ion hopping barrier	1 eV
a_a	Field lowering factor	0.75 nm
$\sigma_{n,tan}$	TaN electric resistivity	220 $\mu\Omega$ cm
$k_{th,tan}$	TaN thermal conductivity (400 K)	4 W m ⁻¹ K ⁻¹
$\sigma_{n,Cu}$	Cu electric resistivity	1.7 $\mu\Omega$ cm
$k_{th,Cu}$	Cu thermal conductivity	401 W m ⁻¹ K ⁻¹

and conditions of this work. The Arrhenius model is used to determine the local electrical conductivity over all the grid points $\forall n \in N$ according to

$$\sigma_n = \sigma_0 \exp\left(-\frac{E_{ac}}{k_B T_n}\right), \quad (1)$$

$$\sigma_0 = e \frac{e D_0}{k_B T_n} N_{ov},$$

where k_B is Boltzmann's constant, e is the electron charge, D_0 is the O ion diffusivity factor, E_{ac} is the activation energy, and σ_0 is the pre-exponential conductance.

The simulation starts from an initial configuration and progresses to a pre-determined final configuration in steps, by either accepting or rejecting permutations in the OV density (N_{ov}) at each step, according to a minimal energy criteria. In order to determine the temperature, potential, and conduction, the charge continuity and steady-state Fourier equations are solved in a self-consistent manner. Since N_{ov} is evaluated through the local conductivity, the Poisson equation is used to account for accumulations of O species by the charge density $\rho_n = -e Z N_o$ (Z is the O ion charge number and ε is the RSL permittivity)

$$\begin{aligned} \nabla \cdot \sigma_n \nabla \varphi_n &= 0, \\ \nabla^2 \varphi_n &= \rho_n / \varepsilon, \\ -\nabla \cdot k_{th,n} \nabla T_n &= \sigma_n |\nabla \varphi_n|^2. \end{aligned} \quad (2)$$

The change in N_o is given by the drift diffusion relation

$$\begin{aligned} \Delta N_{o,n} &= \frac{1}{f_o} \nabla \cdot \left(D_n \nabla N_{o,n} - \frac{e D_n}{k_B T_n} \nabla \varphi_n N_{o,n} \right), \\ D_n &= D_0 \exp\left(-\frac{E_h}{k_B T_n}\right), \end{aligned} \quad (3)$$

where f_o is the vibrational frequency of the O ion. The material dependent values of D_0 and diffusion activation energy E_h are listed in Table I.

The existence of an OV on a grid site yields favorable energy conditions for the migration of Cu.^{15,16} In ST2, where the TE is in direct contact with the RSL, the Cu ion oxidation and reduction probabilities are calculated according to

$$P_n^{o,r} = \exp\left(-\frac{E_{o,r} \pm \frac{1}{2}(a_a e \xi_n + \Delta \varphi_{RSL-M})}{k_B T_n}\right), \quad (4)$$

where a_a is the field lowering factor for generation, ξ_n is the local electric field, $\Delta \varphi_{RSL-M}$ is the RSL-TE work function difference, and $E_{o,r}$ is the activation energy for Cu oxidation and reduction, respectively.¹⁷

ST1, on the other hand, incorporates a TaN based DBL, a material commonly used to prevent Cu migration from routing lines in the IC industry. In this work, a DBL composed of a 2 nm thick TaN thin film is used. The formation of TaNO at the anode interface of resistive memory devices with a metal oxide based RSL was observed by Zhou *et al.*¹⁸ Ou *et al.*¹⁹ demonstrated that plasma treatment may be used to improve TaN barrier performance against Cu diffusion

due to the formation of an interfacial amorphous layer. In addition, the grain size of TaN(O)/TaN films could vary from 2 to 20 nm. In a different publication, Ou *et al.*²⁰ demonstrated that oxygen plasma treatment in ultrathin TaN barrier layer improves the barrier performance by lengthening the grain structures to reduce Cu diffusion.

The DBL thickness is in the order of the TaN grain size and thus grain boundary diffusion is assumed to occur through it. The Arrhenius diffusion model by Lin and Lee²¹ was adopted in this work to calculate the Cu diffusivity as a function of localized temperature and N/Ta ratio (TaN_x) according to the following equation:

$$D_b = \frac{0.661}{\delta} \cdot \left(-\frac{\partial \ln(C)}{\partial y^{6/5}} \right)^{-5/3} \cdot \left(\frac{4D_l}{t} \right)^{1/2}, \quad (5)$$

where D_l and D_b are the lattice and grain boundary diffusion coefficients, respectively. t is the diffusion quasi steady state simulation time step,⁷ and the grain boundary width δ is assumed to be 2 nm. The slopes of $\ln(C)$ vs. $y^{6/5}$ given in the plots for different temperatures¹⁶ are used to determine D_b as a function of temperature.

The temperature dependence of the thermal conductivity in the TaN film was modeled based on the data published in Ref. 22. The thermal oxidation of TaN was modeled according to Brady *et al.*²³ using an activation energy of 1.45 eV at temperatures above 250 °C where fine-grained body-centered cubic, hexagonal close-packed, and face-centered cubic showed the same oxidation kinetics and activation energy.

EXPERIMENTAL DEMONSTRATION

The simulation results presented in this work are compared with the experimental data measured from similar devices. Both structures were constructed starting at a 10-nm-thick TiN BE which was manufactured by means of atomic layer deposition (ALD) and patterned by lithography and reactive-ion etching (RIE). Subsequently, 10 nm thick HfO₂ RSL films were also deposited by ALD and patterned in the

same manner. For ST1, a 2 nm thick TaN DBL was then deposited by ALD. The use of TaN as a barrier layer to construct the DBL was chosen mainly due to its relatively low temperature deposition and low surface mobility. Finally, 100 nm thick Cu TEs were added by electron-beam evaporation at room temperature and patterned through lift-off (both ST1 and ST2).

The initial forming for both structures was carried out using $V_f = 2$ V. A DC endurance test was carried out for both structures for 1000 programming and erase cycles, and the LRS and HRS were determined using a read voltage of 0.3 V.

A schematic configuration and top view of the Cu/TaN/HfO₂/TiN device (ST1) is given in Fig. 1. Both devices discussed in this work have a similar structure and differ only in the existence of the DBL as detailed previously.

RESULTS

The evolution of a CF involves generation, recombination, and hopping of O and OV, which take place on the grid points,⁹ in addition to the migration of TE supplied Cu ions.¹¹ The existence of an OV on a grid site yields favorable energy conditions for the migration of Cu.^{15,16} It was shown

FIG. 1. Schematic description of ST1: (a) device cross sectional view; (b) crossbar structure SEM image; and (c) patterning steps (BE: bottom electrode, AA: active area, and TE: top electrode).

FIG. 2. Schematic illustration of the CFs (highlighted by a blur white triangle) discussed in this work. (a) ST1: a TaN diffusion barrier (thin dashed yellow) between the RSL and the TE results in the formation of a TaNO phase (thick dashed green) with higher blocking properties thus stopping further supply of Cu by the TE to the RSL. (b) ST2: a Cu TE serves as an infinite species reservoir, resulting in a progressively expanding CF during cycling and higher degradation with a continuous reduction of the HRS/LRS separation.

in Ref. 9 that “Hot Spots” (HS), created during the early forming stages within the RSL, serve as agglomeration centers attracting Cu ions to form an initial stem, out of which a CF evolves and grows from the TE toward the HS and eventually to the BE.

The rising RSL temperature combined with O migration toward the TE has a distinct effect on the two simulated structures during CF initial forming as illustrated in Fig. 2. Referring to Fig. 2(a), the depicted structure (ST1) shows that O species have reacted with the TaN layer (marked by a dashed yellow line) to form a sub-stoichiometric tantalum oxynitride TaN_xO_y film ($x + y < 1$, marked by a dashed green line). This formed layer has improved blocking capabilities that further suppress Cu diffusion. The process results in the formation of a thin and narrow CF (highlighted by a blur white triangle). In addition, the lateral widening of the CF during cycling is also restrained. Both processes yield an isomorphic thin and narrow CF that maintains its overall shape, and as a result, the associated electrical properties are more stable. Referring to Fig. 2(b), the depicted structure (ST2) shows that Cu insertion into the RSL is an ongoing process as the device undergoes programming and erase operation cycles. The overall amount of Cu diffusing into the RSL from the TE is much larger than in ST1 and the resulting CF is thus wider. This, in turn, results in a larger divergence in the electrical properties during cycling.

Figure 3 depicts the ST1 simulation results at the end of the initial CF forming for a TE voltage of $V_f = 2.85$ V. The

“formed” criterion is defined to be at an overall device current of 1.25 mA (the calculation method is detailed in Ref. 4) in a similar manner to the current compliance mechanism used in the experimental setups. The resulting density plots for N_{ov} , N_{Cu} , σ_n , and N_{O} are given in Figs. 3(a), 3(b), 3(c), and 3(d), respectively. The calculated low resistance state (LRS) value is ~ 2.3 K Ω . The combination of migrating O species at the TaN-RSL interface with the HS induced high temperature profile of $\sim 450^\circ\text{C}$ [Fig. 3(e)] results in the formation of TaN_xO_y as indicated in Fig. 3(d) by the non-zero concentration of $N_{\text{O,TaN}}$.

Figure 4 depicts the ST1 simulation results at the end of the reset operation for a TE voltage of $V_r = -0.65$ V. The ruptured section is marked by a yellow arrow in Fig. 4(c), where the conductivity was reduced to almost zero. The CF rupture is the result of Cu species migrating back toward the negatively charged TE [Fig. 4(b)] and of OV [Fig. 4(a)] recombining with O [Fig. 4(d)] and driven by both drift and diffusion forces. The higher Cu concentration is evident at the base of the CF in Fig. 4(b) as well as the lower OV concentration in Fig. 4(a) when compared with Fig. 3(a). The $N_{\text{O,TaN}}$ concentration indicating the TaN_xO_y section in Fig. 4(d) remains to be larger than zero thus preventing additional Cu diffusion into the RSL during cycling.

Figure 5 shows the re-forming of the CF using $V_f = 2.85$ V. The previously ruptured section is now re-formed (marked by a yellow arrow) mainly by the creation of additional OV [Fig. 5(a)] and redistribution of Cu from the base

FIG. 3. A CF (marked by a yellow arrow) formed in a Cu/TaN device (ST1) using $V_f = 2.85$ V; (a) N_{ov} , (b) N_{Cu} , (c) conductivity, (d) N_{O} (the TaN layer is highlighted by a yellow dashed line) with a TaN_xO_y film formed by the diffusion of O into the TaN layer, and (e) temperature profile.

FIG. 4. A ruptured CF (ruptured section is marked by a yellow arrow) in a Cu/TaN device (ST1) using $V_f = -0.65$ V; (a) N_{ov} , (b) N_{Cu} , (c) conductivity, and (d) N_o (the TaN layer is highlighted by a yellow dashed line).

of the CF [Fig. 5(b)], where the concentration N_{Cu} was lowered. The addition of new Cu into the base of the CF from the TE is, however, prevented by the TaN_xO_y layer. Some new minor Cu stubs do evolve near the base of the main CF [Fig. 5(b)], followed by lateral widening of the TaN_xO_y within the TaN [Fig. 5(d)] that prevents their further growth. The redistribution of Cu in the CF [Fig. 5(c)] yields a narrower CF, when compared with the freshly formed one [Fig. 2(a)], which in turn leads to the steady small increase in the LRS value with cycling.

Figure 6 shows the concentrations for a re-ruptured CF using a voltage of $V_f = -0.65$ V. The re-ruptured CF section (marked by a yellow arrow) shows once again a close to zero conductivity [Fig. 6(c)]. The TaN_xO_y interfacial layer is now present throughout the entire TaN-RSL interface [Fig. 6(d)] and further Cu diffusion into the RSL is almost completely

suppressed. The size of the previously mentioned Cu stubs is fixed as well [Fig. 6(c)], which prevents the formation of multi CF and helps to stabilize the device's electric parameters.

Figure 7 depicts the ST2 simulation results at the end of the initial CF forming for a TE voltage of $V_f = 2.85$ V. The resulting density plots for N_{ov} , N_{Cu} , σ_n , and N_o are given in Figs. 7(a), 7(b), 7(c), and 7(d), respectively. The calculated low resistance state (LRS) value is once again ~ 2.3 K Ω . Figure 8 shows the species densities in the RSL after the first reset where a ruptured section appears in the CF. Figures 9 and 10 show the same densities in the re-formed and re-ruptured CF after additional set and reset operations, respectively, using the same TE potentials as in the case of ST1.

Since ST2 does not contain a DBL, the formed CF has a wider base [Fig. 7(b)], when compared with the CF base in ST1, which increases in size as the device is ruptured by the

FIG. 5. A cycled CF (marked by a yellow arrow) re-formed in a Cu/TaN device (ST1) using $V_f = 2.85$ V; (a) N_{ov} , (b) N_{Cu} , (c) conductivity, and (d) N_o (the TaN layer is highlighted by a yellow dashed line).

FIG. 6. A cycled CF re-ruptured (ruptured section is marked by a yellow arrow) in a Cu/TaN device (ST1) using $V_f = -0.65$ V; (a) N_{OV} , (b) N_{Cu} , (c) conductivity, and (d) N_O (the TaN layer is highlighted by a yellow dashed line).

migration of Cu toward the TE [Fig. 8(b)] and with cycling due the addition of Cu from the TE [Figs. 9(b) and 10(b)]. This CF lateral widening may account for the higher degradation of electrical properties during cycling as illustrated in Figs. 11 and 12. In addition, the CF of ST2 is based almost entirely on Cu due to OV-O recombination occurring during

cycling [Figs. 9(a) and 10(a)] and thus contains more random variety in its shape which may also contribute to the degradation of electrical properties. On the other hand, the existence of OV along the formed CF path in ST1 [Figs. 4(a) and 5(a)] creates favorable conditions for the migration of Cu along the same path, resulting in an isomorphic CF.

FIG. 7. A CF (marked by a yellow arrow) formed in a Cu (ST2) device using $V_f = 2.85$ V; (a) N_{OV} , (b) N_{Cu} , (c) conductivity, (d) N_O , and (e) temperature profile.

FIG. 8. A ruptured CF (ruptured section is marked by a yellow arrow) in a Cu device (ST2) using $V_r = -0.65$ V; (a) N_{ov} , (b) N_{cu} , (c) conductivity, and (d) N_o .

The experimentally **measured** resistances of the HRS and LRS for ST1 and ST2 devices are summarized in the form of histogram and cumulative distribution function (CDF) plots of Fig. 11, for 1000 set and reset cycles. The mean value (Γ) and standard deviation (Σ) of the HRS and LRS are also presented for both structures. It can be seen that $\Gamma_{HRS,ST1}$ [Fig. 11(c)] is 3 orders of magnitude larger than $\Gamma_{HRS,ST2}$ [Fig. 11(a)] and that $\Gamma_{LRS,ST1}$ [Fig. 11(d)] is in the same order as $\Gamma_{LRS,ST2}$ [Fig. 11(b)]. $\Sigma_{LRS,ST2}$ [Fig. 11(b)] is, however, twice as large as $\Sigma_{LRS,ST1}$ [Fig. 11(d)], which yield more widely dispersed LRS values and degrades the overall reliability.

The ST1 device resulted in much larger endurance than ST2 while maintaining over 3 orders of magnitude separation between the values of the LRS and HRS. Despite the much smaller standard deviation of the HRS for ST2 [$\Sigma_{HRS,ST2}$ in Fig. 11(a)] when compared with that of ST1 [$\Sigma_{HRS,ST1}$ in

Fig. 11(c)], the endurance of ST1 is still considered to be much better. The first reason is the higher mean value of the HRS in ST1 which increases the LRS-HRS separation by 3 more orders of magnitude than that in ST2. The second is that the large distribution is mainly due to even larger HRS resistance values (up to 6 fold the mean value) that increase the said separation even more. Two of the main factors that account for a reliable RRAM device are the LRS-HRS margin and a predictable LRS. The results for ST1 are superior to those of ST2 in both cases.

The **calculated** resistances of the HRS and LRS for ST1 and ST2 devices are summarized in the plots of Fig. 12, for 1000 set and reset cycles with the shaded areas of each plot indicating the distribution of the **measured** resistance as a function of the cycle number. The calculated HRS for ST2 is in ~ 0.7 M Ω [Fig. 12(a)] and the LRS is in the range of 2–5 K Ω [Fig. 12(b)]. The calculated HRS for ST1 is in ~ 10

FIG. 9. A cycled CF (marked by a yellow arrow) re-formed in a Cu device (ST2) using $V_r = 2.85$ V; (a) N_{ov} , (b) N_{cu} , (c) conductivity, and (d) N_o .

FIG. 10. A cycled CF re-ruptured (ruptured section is marked by a yellow arrow) in a Cu device (ST2) using $V_r = -0.65$ V where lateral widening of the CF at the base near the TE is evident; (a) N_{ov} , (b) N_{cu} , (c) conductivity, and (d) N_o .

MΩ [Fig. 12(c)] and the LRS is in the range of 2–2.8 KΩ [Fig. 12(d)].

The quantitative differences between the measured and calculated data shown in Fig. 12 may be attributed to the differences between the presented model and the actual CF. The 3D estimation of the CF resistance is done using an assumption of a circular cross section as mentioned previously. In addition, the model is based on a perfectly planar structure which may not necessarily be the case in the actual device (Fig. 1). Nonetheless, the calculated results are within less than an order of magnitude difference from the measured data in the worst case (LRS of ST2 at forming as shown in Fig. 12). In addition, the calculated results are consistent with the previously published data¹¹ which in turn show a good match to the experimental data.

Both the lower LRS variability and HRS-LRS margin discussed previously are apparent in the simulated results as well and further support the superior reliability attributed to

ST1 over ST2. The larger increase both in the mean value and parametric distribution of the LRS in ST2 [Fig. 12(b)] may be explained by CF lateral widening as discussed previously. The random nature of the algorithm accounts for some of the parametric distribution as well, however, the larger variation seen in Fig. 12(b) when compared with Fig. 12(d), is a good indication that the LRS of ST1 is indeed more stable.

The calculated LRS depicted in Fig. 12 shows a distinctive behavior that may be explained by the simulation results. The LRS of ST1 and ST2 is almost the same at first since the structure of the newly formed CF is relatively similar (Figs. 3 and 7). As the devices are cycled, the LRS of ST2 [Fig. 12(b)] increases in value when compared with ST1 [Fig. 12(d)]. The first reason is the elimination of OV due to a higher temperature profile [Fig. 12(e)] which in turn leads to increased recombination [Figs. 8(a) and 9(a)] and to an

FIG. 11. A histogram and CDF plots of the experimentally measured HRS and LRS during 1000 forming and rupture cycles, Γ and Σ represent the mean value and standard deviation, respectively; ST2: (a) HRS and (b) LRS; ST1: (c) HRS and (d) LRS.

FIG. 12. A plot of the calculated HRS and LRS during 1000 forming and rupture simulation cycles, the shaded areas of each plot indicate the distribution of the measured resistance as a function of the cycle number; ST2: (a) HRS and (b) LRS; ST1: (c) HRS and (d) LRS (inset: y-axis zoom in).

FIG. 13. Experimentally measured forming and resistive switching I-V curves for the structures discussed in this work: (a) ST1 and (b) ST2.

FIG. 14. Experimental DC endurance test at room temperature with a read voltage of 0.3 V for the structures discussed in this work. The plots show the measured device current in the HRS and LRS for both (a) ST1 and (b) ST2.

overall reduction of the conductivity in the CF [Fig. 9(c)]. The second reason is the migration and accumulation of Cu species toward the TE [Figs. 9(b) and 10(b)] resulting in a triangular shaped CF (cone shaped in 3D). The LRS of the CF thus contains a high resistive narrow bottleneck section near the BE. In this manner, the overall LRS in the conventional structure (ST2) increases more with cycling when compared with ST1.

The I-V curves for forming and resistive switching for ST1 and ST2 devices are given in Figs. 13(a) and 13(b),

respectively. The forming voltage is about 2.8 V for ST1 and around 2 V for ST2. The LRS and HRS separation ratio was measured to be roughly 10^4 for both devices at a read voltage of 0.3 V.

The endurance tests for both ST1 and ST2 are depicted in Figs. 14(a) and 14(b), respectively. The distributions in the LRS in ST1 are evidently much smaller than that in ST2 which may indicate more random variation in the formation and rupture of the CF as discussed previously. The HRS yields a read current in the range of $10^{-6} \sim 10^{-10}$ A in ST1

Multimedia

FIG. 15. Simulation progress for the CF formed in ST1 and ST2 as presented in this work ($t=0$: N_{Ov} and N_O starting at a random distribution while $N_{Cu}=0$). Multimedia view: <https://doi.org/10.1063/1.5008727.1>

[Fig. 14(a)] and in the range of $10^{-5} \sim 5 \times 10^{-7}$ A in ST2 [Fig. 14(b)]. The LRS, on the other hand, yields a current of $10^{-3} \sim 10^{-4}$ A for both cases which indicates about the same resistive value. This result is expected and is consistent with the simulation results [Figs. 12(b) and 12(d)], since the physical structure, dimensions, and TE material are similar in both cases. The ST1 device thus maintains a LRS-HRS separation of about 4 orders of magnitude for more than 1000 cycles with very little degradation [Fig. 14(a)], while ST2 shows significant degradation after 500 cycles [Fig. 14(b)].

Figure 15 (Multimedia view) shows the simulation progress for the discussed CF cases and structures and is provided for illustrative purposes.

CONCLUSIONS

The endurance properties of an improved DBL incorporating CBRAM structure are investigated through numerical simulation and compared with a conventional one. The results indicate that the combination of O species migrating toward the TE with a rising RSL temperature during initial CF forming, result in a sub-stoichiometric tantalum oxynitride TaN_xO_y interfacial layer ($x + y < 1$) with higher blocking capabilities that suppresses further Cu insertion beyond this phase. This in turn yields a thin and narrow CF, while at the same time preventing its lateral widening during cycling, a phenomena that is clearly evident in the conventional structure. The endurance of both structures is then evaluated through 1000 programming and erase simulation cycles. The low and high resistive state values for each cycle are calculated, and the analysis reveals that adding the DBL yields lower degradation, which can be attributed to this relatively low amount of Cu. Furthermore, the isomorphic CF displays similar cycling behavior to neural ionic channels while maintaining shape, structure, and electronic properties and may present a promising potential for realizing brain inspired computing platforms. The simulation results show a good match to experimental measurements of similar device structures as well.

ACKNOWLEDGMENTS

The experimental data presented in this work were obtained by the support of the Nano-fabrication Core Lab at King Abdullah University of Science and Technology (KAUST).

- ¹L. O. Chua, "Memristor—Missing circuit element," *IEEE Trans. Circuit Theory* **18**, 507–519 (1971).
- ²L. O. Chua and S. M. Kang, "Memristive devices and systems," *Proc. IEEE* **64**, 209–223 (1976).
- ³T. Y. Tseng and S. M. Sze, *Nonvolatile Memories: Materials, Devices, and Applications* (American Scientific Publishers, CA, USA, 2012), Vol. 1, p. 1.
- ⁴D. B. Strukov *et al.*, in *ISCAS*, 2010, p. 1967.
- ⁵H.-S. P. Wong *et al.*, *Nano Lett.* **12**, 2179 (2012).
- ⁶J. J. Yang, D. B. Strukov, and D. R. Stewart, *Nat. Nanotechnol.* **8**, 13 (2013).
- ⁷H.-S. P. Wong, H. Y. Lee, S. Yu, Y. S. Chen, Y. Wu, P. S. Chen, B. Lee, F. T. Chen, and M. J. Tsai, "Metal-oxide RRAM," *Proc. IEEE* **100**(6), 1951–1970 (2012).
- ⁸K. Aratani *et al.*, "A novel resistance memory with high scalability and nanosecond switching," *IEEE Int. Electron Devices Meet.* **2007**, 783–786.
- ⁹D. Berco and T. Y. Tseng, *J. Comput. Electron.* **15**(2), 577–585 (2015).
- ¹⁰D. Berco and T.-Y. Tseng, *Appl. Phys. Lett.* **107**, 253504 (2015).
- ¹¹D. Berco and T. Y. Tseng, *J. Comput. Electron.* **15**(2), 586–594 (2015).
- ¹²D. Berco and T.-Y. Tseng, *AIP Adv.* **6**, 025212 (2016).
- ¹³U. Chand, T.-Y. Tseng *et al.*, *Appl. Phys. Lett.* **106**(16), 153502 (2015).
- ¹⁴U. Chand, T.-Y. Tseng *et al.*, *J. Appl. Phys.* **117**(18), 184105 (2015).
- ¹⁵J. Guy, G. Molas, P. Blaise, C. Carabasse, M. Bernard, A. Roule, G. Le Carval, V. Sousa, H. Grampeix, V. Delaye, A. Toffoli, J. Cluzel, P. Brianceau, O. Pollet, V. Balan, S. Barraud, O. Cueto, G. Ghibaudo, F. Clermidy, B. De Salvo, and L. Perniola, *IEEE Int. Electron Devices Meet.* **2014**, 6.5.1–6.5.4.
- ¹⁶G. Molas, E. Vianello, F. Dahmani, M. Barci, P. Blaise, J. Guy, A. Toffoli, M. Bernard, A. Roule, F. Pierre1, C. Licitra, B. De Salvo, and L. Perniola, *IEEE Int. Electron Devices Meet.* **2014**, 6.1.1–6.1.4.
- ¹⁷W. M. Haynes, *CRC Handbook of Chemistry and Physics*, 91st ed. (CRC Press, Boca Raton, FL, 2010), pp. 5-16–5-17.
- ¹⁸P. Zhou, M. Yin, H. J. Wan, H. B. Lu, T. A. Tang, and Y. Y. Lin, *Appl. Phys. Lett.* **94**, 053510 (2009).
- ¹⁹K.-L. Ou, W.-F. Wu, C.-P. Chou, S.-Y. Chiou, and C.-C. Wu, *JVSTB* **20**, 2154 (2002).
- ²⁰K.-L. Ou, M.-H. Lin, and S.-Y. Chiou, *Electrochem. Solid-State Lett.* **7**(11), G272–G275 (2004).
- ²¹J.-C. Lin and C. Lee, *J. Electrochem. Soc.* **146**(9), 3466–3471 (1999).
- ²²E. Bozorg-Grayeli *et al.*, *Appl. Phys. Lett.* **99**, 261906 (2011).
- ²³D. P. Brady, F. N. Fuss, and D. Gerstenberg, *Thin Solid Films* **66**(3), 287–302 (1980).