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1 Resistance controllability and variability improvement in a TaO_x-based 2 resistive memory for multilevel storage application

AQ1 3 A. Prakash,^{1,a), b)} D. Deleruyelle,^{2,a)} J. Song,¹ M. Bocquet,² and H. Hwang^{1,b)}

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8 In order to obtain reliable multilevel cell (MLC) characteristics, resistance controllability between
9 the different resistance levels is required especially in resistive random access memory (RRAM),
10 which is prone to resistance variability mainly due to its intrinsic random nature of defect generation
11 and filament formation. In this study, we have thoroughly investigated the multilevel resistance
12 variability in a TaO_x-based nanoscale (<30 nm) RRAM operated in MLC mode. It is found that the
13 resistance variability not only depends on the conductive filament size but also is a strong function
14 of oxygen vacancy concentration in it. Based on the gained insights through experimental
15 observations and simulation, it is suggested that forming thinner but denser conductive filament may
16 greatly improve the temporal resistance variability even at low operation current despite the
17 inherent stochastic nature of resistance switching process. © 2015 AIP Publishing LLC.
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18 Resistive random access memory (RRAM) is one of the
19 most promising next generation memory candidates espe-
20 cially for storage class memory applications due to its scal-
21 ability, structural simplicity, and backend process compatible
22 stackability.¹⁻³ In addition, TaO_x as a RRAM switching
23 material have been found to be of commercial interest due to
24 its superior material property as well as memory perform-
25 ance.³⁻⁶ To fulfill the increasing demand for low cost and
26 higher density memory, the most obvious way to increase the
27 storage capacity is by decreasing the physical size of the
28 device to a nanoscale dimensions, and in RRAM the scalabil-
29 ity down to 10 nm has been demonstrated.^{7,8} Other interesting
30 approach is to stack the devices 3-dimensionally, for which
31 two feasible architectures of crossbar and vertical RRAM are
32 reported.⁹ However, both the methods require complex exper-
33 imental processes and may suffer from other limitations.
34 Another alternative and simpler way to increase storage
35 density is to use multilevel cell (MLC) storage technology, in
36 which more than one bit per cell can be stored without further
37 decreasing the physical device size.^{10,11} However, precise
38 control over the resistance of the different resistance levels
39 should be assured for reliable MLC operation especially for
40 RRAM, which is known to suffer from variability and reli-
41 ability issues due to certain randomness in its switching
42 process.¹²⁻¹⁴

AQ2 43 In RRAM, MLC characteristics can be obtained by vari-
44 ous methods such as by varying the switching current (or
45 compliance current), where different levels of low resistance
46 state (LRS) can be obtained due to change in the total num-
47 ber of constituent defects of the conductive filament (CF) or
48 by controlling the maximum reset voltage during reset

operation which can result different levels of high resistance
state (HRS) due to variation in the gap between CF tip and
metal electrode.^{10,11} MLC behavior by varying the program/
erase pulse width is also reported.¹¹ Although MLC proper-
ties can easily be obtained in RRAM, its successful imple-
mentation is dictated mainly by the ability to precisely
control the resistance margin between the two resistance
levels. One of the most critical factors, which can degrade
this margin, is the switching cycle-to-cycle variability.
Therefore, the feasible approaches to minimize it should be
delved into through the enhanced understanding of the
responsible factors. The origin of this variability is mainly
related to the change in the total content of the oxygen
vacancy defects present in the CF after each switching
event.¹² This compositional variation in the conductive chan-
nel leads to the conductivity fluctuation. More fundamen-
tally, it can be understood in terms of having different
activation energy of defect migration due to the random
location and different surroundings of the defects with in the
conductive channel.

In this study, we aim to develop enhanced understanding
on the variability issue through experimental observation
and simulation studies and correlate it with the CF size and
defect concentration in it. For this, we fabricated nanoscaled
(<30 nm) RRAM devices with TaO_x as a switching material.
Based on the analysis of experimental observations and sim-
ulation, a possible way to minimize the variability between
resistance levels for reliable MLC operation is proposed.
The results show that the temporal resistance variability may
greatly be improved even at low switching current by form-
ing denser filament with smaller size. Monte-Carlo calcula-
tions show good agreement with the experimental data.

The device was fabricated on a patterned substrate with
30 nm TiN bottom electrode (BE) in a planner type device
structure [see the inset of Fig. 1(a)]. The TiN electrode was
deposited by CVD and its sides were protected by 100 nm

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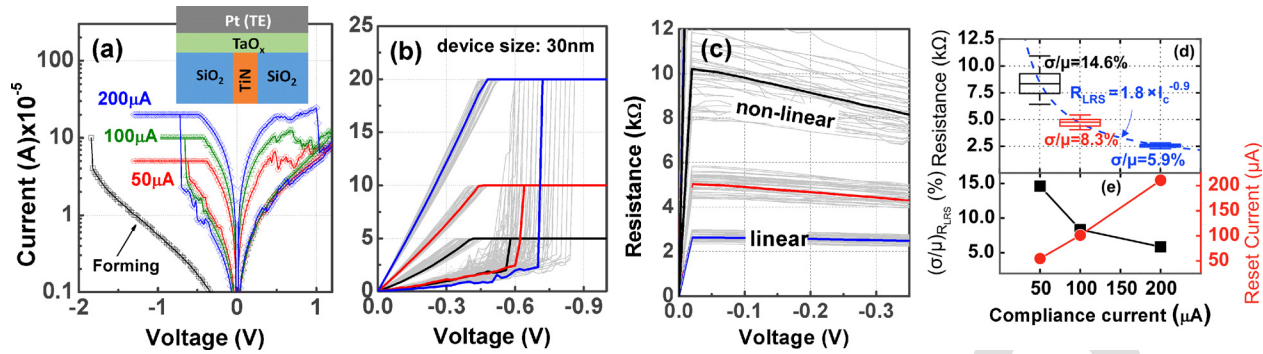


FIG. 1. (a) Forming and resistive switching behavior of fabricated 30 nm sized TaO_x-based resistive memory device with MLC characteristics. (b) Multiple MLC current-voltage (I-V) curves of all the LRS levels showing stable MLC characteristics. (c) Corresponding resistance-voltage (R-V) plot of all the low resistance levels confirming nonlinear conduction at smaller switching current. (d) Resistance of LRS and its distribution as a function of I_C. Both decrease with increasing I_C. (e) Evolution of LRS variability (σ/μ) and reset current (I_{reset}) with I_C.

thick SiO₂ layer. Tantalum oxide (TaO_x) switching layer of ~10 nm was deposited by reactive sputtering using Ta metal target and Ar+O₂ gases at room temperature. The rf power and working pressure were 100 W and 3 mTorr, respectively. Finally, inert (Pt) top electrode (TE) was sputter deposited on TaO_x switching layer. For electrical measurements, voltage was applied on TE and BE was connected to the ground.

Multilevel cell operation and resistance variability analysis of fabricated 30 nm size RRAM device is presented in Fig. 1. First, the device was formed by applying a negative voltage on Pt TE, as shown in Fig. 1(a). Small voltage of < -2.0 V was sufficient to form the device. After forming, the device exhibits resistive switching behavior and MLC characteristics by varying the switching current, as shown in Figs. 1(a) and 1(b). Multiple cycles for each level have been shown to ensure the stable resistance switching. Three different levels of LRS were obtained with same HRS, which can be used in 2-bit per cell storage. In addition, it was observed that the LRS current-voltage (I-V) gradually shifted from an ohmic to a nonlinear behavior as current compliance (I_C) was decreased down to 50 μA (Fig. 1(c)) due to decreased number of vacancy defects in the CF leading to the change in the current conduction mechanism. Simultaneously, the resistance margin between the resistance levels is reduced because of cycle-to-cycle resistance fluctuations. The resistance distribution of 50 consecutive cycles for each compliance is shown in Fig. 1(d), where the highest variability of 14.6% is observed for I_C of 50 μA. The read voltage was 0.25 V. Furthermore, the resistance of LRS (R_{LRS}) decreased with increasing I_C [Fig. 1(d)]. The slope of R_{LRS} vs I_C was found to be -0.9, which is close to the reported value of -1.¹² On the other hand, maximum reset current (I_{reset}) increases linearly and coefficient of variation (σ/μ) or variability of R_{LRS} decreases with increasing I_C, as shown in Fig. 1(e). The decrease in the variability is due to the increase in the number of oxygen vacancy defects in the CF with increasing I_C. Considering the dependence of LRS I-V nonlinearity on I_C and variability behavior, it can be said that by increasing I_C, both CF size and oxygen vacancy defect density in it may increase. In other words, R_{LRS} depends on filament size and its electrical conductivity.

The different electrical conduction behavior (linear/nonlinear) of the LRS I-V curves observed at different

switching currents [Fig. 1(c)] can be understood by the oxygen vacancy concentration (n_{Vo}) falling under a threshold value (n_{TAC}), which is the minimum concentration of oxygen vacancies below which trap assisted conduction (TAC) dominates and I-V becomes nonlinear, while above this concentration conduction is linear (ohmic).¹⁵⁻¹⁸ This can be accounted by the following equation:

$$\sigma_e(n_{\text{Vo}}) = \beta \times n_{\text{Vo}} \times \exp\left(-\frac{E_a(n_{\text{Vo}})}{kT}\right), \quad (1)$$

$$\text{with } E_a(n_{\text{Vo}}) = E_{a0} \times f(n_{\text{Vo}}), \quad (2)$$

where σ_e is the electrical conductivity, E_a is the activation energy, $\beta = 1 \times 10^{-23} \text{ S m}^{-1}$, k = Boltzmann constant, T = absolute temperature, and $E_{a0} = 0.4 \text{ eV}$. This approach has already been employed to describe resistance switching phenomena or early data-retention failure in RRAM employing either HfO_x^{15,16} or TaO_x.^{17,18} The corresponding plot of σ_e as a function of n_{Vo} is shown in Fig. 2. In the previous studies,¹⁵⁻¹⁸ a piecewise linear and decreasing function f [Eq. (2)] was employed to account for the decrease of conduction activation energy at increasing defect concentrations which, in turns, increases σ_e in the CF region. Although a proper theoretical justification is still

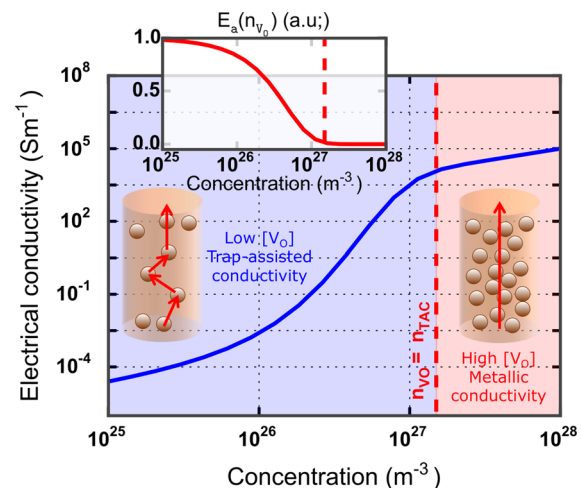


FIG. 2. Calculated electrical conductivity as function of oxygen vacancy concentration [Eq. (1)]. Inset shows the evolution of the activation energy with respect to n_{Vo} .

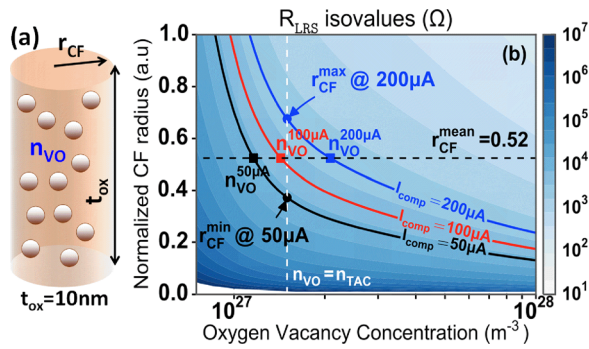


FIG. 3. (a) Schematic description of the conductive filament. (b) Maps of R_{LRS} as function of conductive filament radius and oxygen vacancy concentration. LRS isolines correspond to experimental mean LRS resistance values at different I_C .

lacking, this first-order approximation showed satisfactorily and quantitative agreement with data.^{15,17,18} In this work, we employed a sigmoid function [see the inset of Fig. 2] rather than a piecewise linear function to describe the decreasing E_a with n_{VO} . Although such a dependency is only justified as a piecewise decreasing function, it offers the numerical advantage of being indefinitely differentiable in the whole range of n_{VO} to conduct further numerical or analytical studies. In the following, a value of n_{TAC} of $1.5 \times 10^{27} \text{ m}^{-3}$ was employed (marked by vertical dashed line in Fig. 2), which corresponds to an inter-distance below 1 nm between oxygen vacancies. Considering the R_{LRS} equal to the resistance of CF (R_{CF}) in LRS and cylindrical filament [Fig. 3(a)], R_{CF} can be written as

$$R_{LRS} \approx R_{CF} = \left(\frac{1}{\sigma_e(n_{VO})} \right) \times \left(\frac{t_{ox}}{\pi \times r_{CF}^2} \right), \quad (3)$$

where t_{ox} is the oxide thickness ($= 10 \text{ nm}$) and r_{CF} is the conductive filament radius. Figure 3(b) shows the R_{LRS} isovalue curves of all the resistance levels as a function of CF radius and oxygen vacancy concentration (n_{VO}). The n_{VO} equal to n_{TAC} is also shown by a dashed line. The dependence of R_{LRS} versus the vacancy concentration is intensified as the

concentration falls below n_{TAC} (i.e., non-ohmic regime) as indicated by sharper R_{LRS} isovalues [Fig. 3(b)]. As vacancy generation and filament formation are intrinsically random in RRAM, vacancy concentration change in the filament is inevitable during cycle-to-cycle switching. Consequently, small changes in n_{VO} may introduce large variability in the resistance of multilevel, in particular, when $n_{VO} < n_{TAC}$ as observed in the experimental results [Fig. 1]. It is interesting to note that LRS variability not only depends on the filament radius (size) but also is strongly dictated by vacancy concentration and fluctuation in the filament. Interestingly, the graph also shows that the different topologies of filament (i.e., r_{CF} and n_{VO}) can account for the experimental mean R_{LRS} values obtained at different I_C [Fig. 3]. Given the non-linearity (respectively, linearity) of I-V curves at $50 \mu\text{A}$ (respectively, $200 \mu\text{A}$), r_{CF} can be reasonably assumed to be higher (respectively, lower) than r_{CF}^{min} (respectively, r_{CF}^{max}) to account for non-ohmic (respectively, ohmic) I-V.

To better understand the effect of vacancy concentration on the variability, let us assume a mean filament radius (r_{CF}^{mean}) in between r_{CF}^{min} and r_{CF}^{max} , as shown in Fig. 3(b) by horizontal dashed line. Three n_{VO} can now be obtained on the R_{LRS} isovalue curves corresponding to switching currents of 50 , 100 , and $200 \mu\text{A}$. Monte-Carlo calculations have been performed to reproduce the experimental LRS resistance distribution assuming a Gaussian distribution of n_{VO} in Eq. (3). Results are presented in Figs. 4(a)–4(c). A pretty close match between the experimental variability data and calculations has been achieved using the values given in Figs. 4(d) and 4(e). It is interesting to note that a small (i.e., $< 5\%$) variation in n_{VO} can result in a large (14.6%) variability in the resistance for $50 \mu\text{A}$ I_C and a 5.9% (smaller) resistance variation for $200 \mu\text{A}$ I_C . This underlines that the resistance variability is very sensitive to the n_{VO} variation in the CF when it operates in the nonlinear region where electrical conductivity strongly depends on n_{VO} . Conduction activation energies for each computed n_{VO} are in the range between 0.8 meV (for $I_C = 200 \mu\text{A}$) and 16.7 meV (for $I_C = 50 \mu\text{A}$) [Fig. 4(e)]. E_a values are in the same order of magnitude than those already reported by Kim *et al.*^{17,18} Figure 5 suggests possible way to

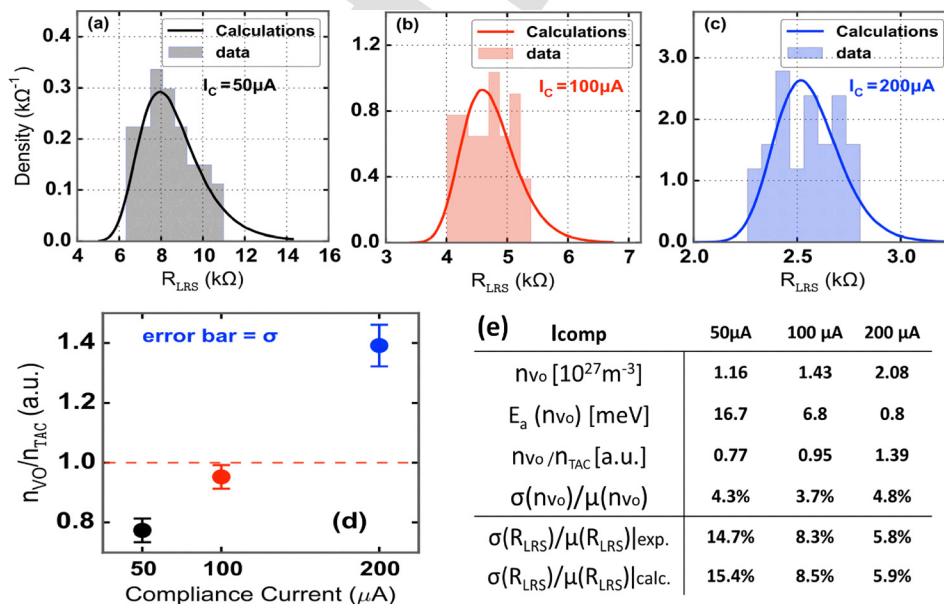


FIG. 4. (a)–(c) Experimental normalized resistance distributions (histograms) and computed density (lines) for each I_C . Densities were computed using mean values and standard deviation of n_{VO} presented in (d). Table (e) is a summary of the experimental and calculation results.

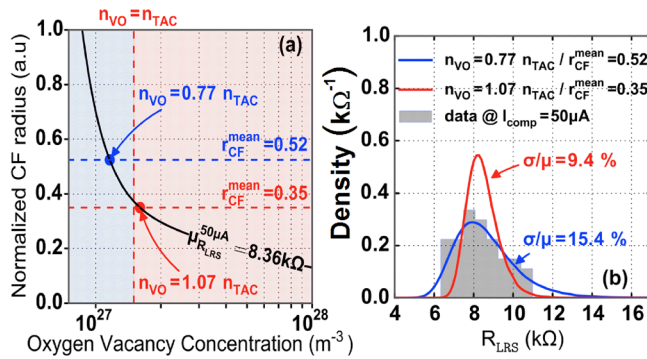


FIG. 5. (a) R_{LRS} isoline for $I_C = 50 \mu\text{A}$. Shaded regions correspond to trap-assisted (left side) and ohmic (right side) transport regions. (b) Experimental (histogram) and computed resistance distribution (lines) using the same relative variation of n_{VO} for different n_{VO}/n_{TAC} values. It is shown that $n_{VO}/n_{TAC} > 1$ allows for smaller relative variation of R_{LRS} .

minimize the resistance variations even at low I_C . It is possible to improve the variations if we can shift to operate in the linear region instead of nonlinear region, as shown in Fig. 5(a). This can be achieved by generating smaller size filament with higher defect concentration. Calculations show that a tighter distribution with 8% variability can be achieved even at low I_C of $50 \mu\text{A}$ if n_{VO}/n_{TAC} is increased from 0.77 to 1.07 in conjunction with r_{CF}^{mean} decreased from 0.52 to 0.35 [Fig. 5(b)]. Therefore, forming denser but thinner filament may remarkably improve the resistance fluctuation even at low operation current. Various techniques such as two step forming, material engineering, and different forming conditions can be exploited to generate different defect density filament.⁶

In this work, multilevel resistance variability was investigated on a nanoscale TaO_x -based RRAM operating in MLC mode by means of a theoretical approach assuming that non-linearity observed at low I_C is related to trap assisted conduction resulting from low concentration of oxygen vacancy. It was found that the resistance variability not only depends on the conductive filament size but also is a strong function of oxygen vacancy concentration and fluctuation, which is supported by Monte-Carlo calculations of LRS distributions at various compliance currents. Based on this study, it is suggested that LRS variability can be improved

even at low I_C by forming denser CF in terms of oxygen vacancy by means of material engineering or dedicated forming techniques.

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