AIP | Applied Physics Letters

The conical shape filament growth model in unipolar resistance switching of TiO 2 thin film

Kyung Min Kim and Cheol Seong Hwang

Citation: Applied Physics Letters **94**, 122109 (2009); doi: 10.1063/1.3108088
View online: http://dx.doi.org/10.1063/1.3108088
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/94/12?ver=pdfcov
Published by the AIP Publishing

The conical shape filament growth model in unipolar resistance switching of TiO_2 thin film

Kyung Min Kim and Cheol Seong Hwang[a)]
Department of Materials Science and Engineering and Inter-university Semiconductor Research Center, Seoul National University, Seoul 151-744, Republic of Korea

(Received 9 January 2009; accepted 8 March 2009; published online 26 March 2009)

This study examined the relationship between the conducting filament resistance and reset voltage during the resistance switching of TiO_2 thin films assuming a filament with a conical shape. There was a critical resistance ($\sim 20\ \Omega$) of the set state above and below which the filament responded differently in response to the current. Maintaining a higher set state resistance was more beneficial in achieving a more uniform reset voltage. This filament model coincides well with the localized switching behavior and the recently microscopically observed filament shape. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3108088]

Recently, resistance switching (RS) phenomena in a variety of thin film materials have been actively studied in order to apply them to the next generation nonvolatile memory devices. Among the various materials, *n*-type TiO_2 and *p*-type NiO have been studied extensively.[1,2] The unipolar RS process proceeds by applying the same bias polarity and has been understood from the formation [set switching from a high resistance state (HRS) to a low resistance state (LRS)] and rupture (reset switching from the LRS to HRS) of percolated conduction channels (called filaments) in these oxides. One of the problems with RS materials is the nonuniform switching parameters such as the set and reset voltages (V_{set} and V_{reset}) as well as the accompanying resistances (R_{set} and R_{reset}), even though the R_{reset}/R_{set} ratio is sufficiently large. Chae *et al.*[3] recently proposed a statistical model of RS using the random circuit breaker hypothesis. They further refined this idea to explain the randomized reset process in NiO using the percolating cluster model with the characteristic length.[4] Although these models explained the random variations in the RS parameters in a statistical manner, this may not necessarily be the case for unipolar RS in TiO_2. This is because TiO_2 exhibits anode interface localized switching (only in the 3–10 nm region near the anode),[5] which cannot be accounted for by the percolating cluster model. Therefore, in this study, a new model that properly explains the RS behavior in TiO_2 is proposed by interpreting filament rupture, of which the shape is assumed to be conical, along with Joule heat generation and dissipation. The authors recently observed the filament shape by high-resolution transmission electron microscopy.[6] The filament consisted of Ti_5O_9 (or Ti_4O_7 and their mixtures that are all metallic conductors) with a diameter of $\sim 5-10$ nm directly connecting the top and bottom electrodes.[6]

The preparation of a Pt/40-nm-thick TiO_2/Pt structured sample and method for RS of the sample is reported in detail elsewhere.[5–9] RS was tested using a semiconductor parameter analyzer (HP 4145B) in voltage sweep mode with the proper setting of the compliance current (I_{comp}). The randomness of the switching parameters, particularly V_{reset} and R_{set}, was due mainly to the random transient of the current (charge) flow when the compliance current was reached during the set process.

The inset in Fig. 1(a) shows the assumed conical shape of the conducting filament, where the radius decreases (r at the top interface is fixed and radius at the bottom interface is ar, where the parameter a is <1) from the top electrode interface (cathode) to the bottom electrode interface (anode). With this model, the location where the peak temperature appears during the reset process could be estimated. Here, the reset process was understood to fill in the oxygen vacancies or transform the metallic Ti_5O_9 phase to an insulating TiO_2 phase by supplying the local region of the filament near the bottom interface with oxygen ions that had electromigrated from the cathode. The migration of oxygen ions is a thermally activated process. Hence, the region with the highest temperature has the highest probability of filament breaking (reset).[10]

In order to estimate the local temperature along the direction of the filaments, it is essential to calculate the heat produced by current flow (the filaments are metallic conductors) as well as the heat dissipated to the nearby areas through the boundary between the filament and matrix. Due to the narrow shape of the filaments, the heat losses to the electrodes can be ignored. Heat transfer between each segment of the filament with a thickness of δd (shown by the ring in the figure) can also be disregarded because they must also be small.

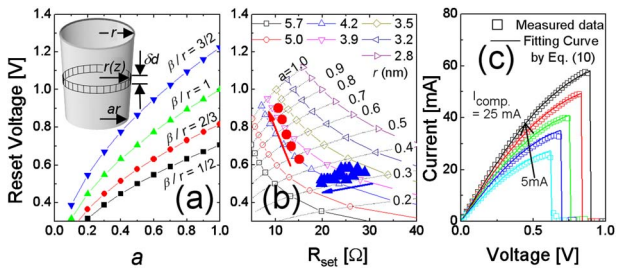

FIG. 1. (Color online) (a) Change in V_{reset} as a function of β/r. The inset shows the assumed conical shape of the conducting filament. (b) The calculated correlation between the R_{set} and V_{reset} with r and a as variables. The measured data from the single Pt/TiO_2/Pt cell and serially connected Pt/TiO_2/Pt/TiO_2/Pt cell are included as red circles and triangles, respectively. (c) The measured *I-V* curves (square) and lines of best fit (solid line).

a)Electronic mail: cheolsh@snu.ac.kr.

The resistance of a conical shaped filament (R_{filament}) is given by the following:

$$R_{\text{filament}} = \left(\frac{1}{a}\right)\frac{\rho d}{\pi r^2}, \quad (a < 1), \tag{1}$$

where ρ is the resistivity of the filament and d is the film thickness. The current flow through a filament can be obtained by Ohm's law ($I = V/R_{\text{filament}}$). Although I is determined by R_{filament}, the local heat generation is proportional to the local current density and local resistance of the element δd. Therefore, the largest heat must be generated at the narrowest point. However, heat dissipation through the boundary should be also highest due to the highest surface to volume ratio there.

Under a steady state, the rate of heat transfer (dQ_{net}/dt) across the filament boundary becomes 0, where Q_{net} and t are the net heat ($Q_{\text{net}} = Q_{\text{gen}} + Q_{\text{dis}}$, where Q_{gen} and Q_{dis} are the generated and dissipated heat, respectively) and time, respectively. The rate of heat generation (dQ_{gen}/dt) is $I^2R[r(z)]$, where $R[r(z)]$ is the local resistance at z with a radius of $r(z)$. Here, $R[r(z)] = \rho\delta d/\pi r(z)^2$. Therefore, dQ_{gen}/dt can be expressed as

$$\frac{dQ_{\text{gen}}}{dt} = I^2R[r(z)] = \frac{V^2}{\left[\left(\frac{1}{a}\right)\frac{\rho d}{\pi r^2}\right]^2}\frac{\rho\delta d}{\pi r(z)^2}. \tag{2}$$

Considering the assumption that heat dissipation occurs only at the filament boundary, the rate of heat dissipation (dQ_{dis}/dt) is $-kA(dT/dx)$, where k is the thermal conductivity of TiO_2, $A = 2\pi r(z)\delta d$, and dT/dx is the temperature gradient at the filament boundary. Thus,

$$\frac{dQ_{\text{dis}}}{dt} = -k(2\pi r(z)\delta d)\left(\frac{dT}{dx}\right) \tag{3}$$

and

$$\frac{Q_{\text{net}}}{dt} = \frac{Q_{\text{gen}}}{dt} + \frac{Q_{\text{dis}}}{dt} = 0. \tag{4}$$

Therefore,

$$\frac{dT}{dx} = \frac{V^2r^4}{\left(\frac{1}{a}\right)^2(2k\rho d^2)r(z)^3}. \tag{5}$$

In the region near the filament, the temperature gradient increases with increasing local temperature because the temperature of the region far from the filament is maintained at room temperature. Equation (5) shows that dT/dx is proportional to $r(z)^{-3}$ for the other given variables. Therefore, the local temperature at that point increases with decreasing $r(z)$. This suggests that the increase in heat generation by current crowding overcompensates for the heat dissipation by the larger surface area to volume ratio. From Eq. (5),

$$V = \sqrt{\left(\frac{1}{a}\right)^2\beta\frac{r(z)^3}{r^4}}, \quad \left(\beta = 2k\rho d^2\frac{dT}{dx}\right). \tag{6}$$

Since the overall reset process must be controlled by the reset of the weakest point along the filament, the V of the smallest $r(z)$ represents the overall V_{reset}. Here, the minimum $r(z)$ is ar so that

$$V_{\text{reset}} = a^{1/2}(\beta/r)^{1/2}, \quad \left(\beta = 2k\rho d^2\frac{dT}{dx}\right). \tag{7}$$

Even without the complicated thermal simulation, Eq. (7) shows that the V_{reset} is inversely proportional to $r^{1/2}$.

The actual calculation of V_{reset} requires the values of ρ, k, and dT/dx. Although determining these values for Ti_5O_9 or Ti_4O_7 phase is difficult, the proper range of β/r and a can be estimated from the measured V_{reset} being ~0.5–1.0 V. Figure 1(a) shows the change in V_{reset} as a function of a according to Eq. (7) with several numerical values of β/r. The figure suggests that the proper β/r values range from 1/2 to 3/2.

The R_{filament} and V_{reset} can be calculated for a given a and r using Eqs. (1) and (7), respectively. Hence, the correlation between R_{filament} and V_{reset} can be achieved as a function of a and r using ρ of $Ti_5O_9 \sim 2\times10^{-5}$ Ω m near room temperature[11] and $\beta = 3.5\times10^{-9}$ V^2 m. The value of β was not calculated by Eq. (7) and the material parameters because of the unknown dT/dx. On the other hand, this value can be estimated from the line of best fit of the current-voltage (I-V) curves as will be shown later. The dT/dx was calculated to be $\sim4.6\times10^9$ K m^{-1} using the β value and thermal conductivity of TiO_2 ($k = 11.7$ W m^{-1} K^{-1}). This appears to be reasonable considering a filament temperature of several hundred degrees and nanometer size of the filament.

The measured R values from the pad I-V experiment (R_{set} at $V = 0.1$ V) of the set state sample range from 10 to 30 Ω. There are many parallel filaments in a single pad that simultaneously conduct the electricity during the R measurements. Therefore, the R_{filament} of a single filament should be calculated by multiplying the measured R_{set} value with the average number of the filaments. There are variations in the filament density on the pad area (100×100 μm^2) depending on the switching conditions (~1000–3000 ea/100×100 μm^2). The specific conductivity and diameter of each filament also vary so that only an approximate filament density (~2000 ea/100×100 μm^2) was used for the calculation of a typical R_{filament} value of one filament. The calculated diameter of the filaments shown in Fig. 1(b) could be varied by ~5%–10%. Figure 1(b) shows the calculated correlation between the R_{set} and V_{reset} by the open symbols with r and a as variables. It should be noted that R_{filament} is 2000 times larger than R_{set} shown in Fig. 1(b).

The experimental data from the single layer TiO_2 samples are included in Fig. 1(b) as red circles. Figure 1(c) shows the I-V curves for those data points, which indicates that V_{reset} in a TiO_2 film increases with decreasing R_{set}. For the previous set experiments, I_{comp} was changed from 5 to 25 mA in 5 mA interval. Figure 1(b) shows that the filament becomes more cylindrical (less conical, a increases) and its radius increases slightly, which is accompanied by a concomitant increase in V_{reset} with decreasing R_{set}.

The reset I-V curves could be simulated accurately by assuming that R_{set} increases as the voltage approaches V_{reset} and the filament temperature increases (the filament is metal). This was also acknowledged by Russo *et al.*[12] for NiO. The R_{set} as a function of V and the I-V curve are given by Eqs. (8) and (9), respectively,

$$R_{\text{set}}(V) = R_0[1 + \gamma(\Delta T)], \tag{8}$$

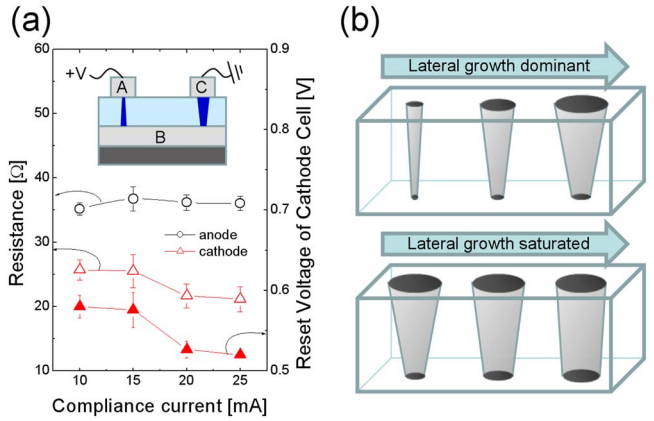

FIG. 2. (Color online) (a) RS results of a serially connected $Pt/TiO_2/Pt/TiO_2/Pt$ cell. The inset shows a serially connected structure and an illustration of the filament shape of each cell. (b) Schematic diagram of the evolution of the filament shape according to I_{comp} (or $R_{filament}$).

$$I = \frac{V}{R_{set}(V)} = \frac{V}{R_0[1+\gamma(\Delta T)]}, \tag{9}$$

where R_0 is the filament resistance at room temperature, γ is the temperature coefficient of resistance, and ΔT is the temperature change in the filament. For steady-state heat conduction at the filament boundary, $dT/dx = \Delta T/\Delta x$. From Eq. (5),

$$I = \frac{V}{R_0[1+\chi(a^{-1}V^2 r)]}, \quad (\chi = \gamma\Delta x/2k\rho d^2). \tag{10}$$

Using R_0 and a in Fig. 1(b) and the material constants, the solid lines of best fit according to Eq. (10) in Fig. 1(c) exactly matched the experimental data. This is a completely different interpretation of the nonlinear *I-V* curves of the reset process from that reported by Lee *et al.*[4] However, the V_{reset} versus R_{set} behavior is similar to the NiO case where the resistance is lower than the critical resistance ($R_o < R_{co}$) in Ref. 4. The following shows another interesting coincidence between this work and the report by Lee *et al.* for the cases of $R_o > R_{co}$.

The RS of a serially connected $Pt/TiO_2/Pt/TiO_2/Pt$ cell, as shown in the inset of Fig. 2(a), was measured and the results are shown in Fig. 2(a). Similar experiments and interpretation were reported previously.[5] In these measurements, the set operation of a serially connected structure was performed by connecting the top electrode "C" to the ground (C is cathode) and applying a positive voltage to the top electrode "A" (A is anode). For this set operation, the I_{comp} was changed from 10 to 25 mA in 5 mA interval. The experiments were carried out five times under each condition for reliability. Every RS was carried from the fresh state to exclude any potential pre-existing filament effect. Fresh state means that those points were not electrically measured previously. The set state resistance between the bottom electrode (denoted as "B") and A ($R_{A\text{-}B}$) was always higher than the resistance between B and C, ($R_{C\text{-}B}$), as shown in Fig. 2(a), which suggests that the filaments formed near the anode are always weaker than those near the cathode. The $R_{A\text{-}B}$ is almost independent of the I_{comp}, whereas the $R_{C\text{-}B}$ decreases with increasing I_{comp}. After measuring $R_{C\text{-}B}$, the reset of a single C-B cell was performed and its V_{reset} was measured, which is also included in Fig. 2(a). Interestingly, in this case, as $R_{C\text{-}B}$ decreases, V_{reset} of the cathode cell also decreases, which is in contrast to the results shown in Fig. 1(c). This corresponds to the cases of $R_o > R_{co}$ in Ref. 4. The thermal simulation results reported by Russo *et al.*[12] based on filaments with a cylindrical shape in NiO are also consistent with this result. A different correlation between the filament shape and $R_{C\text{-}B}$ compared to the cases shown in Fig. 1(c) was found when the relationship between $R_{C\text{-}B}$ and V_{reset} was included in Fig. 1(b) as blue triangles. In this case, the decrease in $R_{C\text{-}B}$ is related to the increase in r with an almost fixed a (in this case, $a \sim 0.35$). This suggests that in this R_{set} ($R_{C\text{-}B}$) range, the decrease in R_{set} is accomplished by thickening of the filament with a fixed a value. However, as the R_{set} decreases further to $< \sim 20\ \Omega$, a decrease in R_{set} (also $R_{filament}$) is accomplished by mainly changing its shape from a conical one to a more cylindrical one. The critical resistance of $R_{set} \sim 20\ \Omega$ is similar to that in the percolation model for NiO ($\sim 25\ \Omega$).[4] Fig. 2(b) gives a schematic diagram showing the evolution of the filament shape according to R_{set}. When the filament was thin (due to the generally smaller I_{comp}) so that R_{set} was $> \sim 20\ \Omega$ (or $R_{filament} > 40\,000\ \Omega$), both the top and bottom of the filament grew laterally with an almost constant top to bottom radius ratio when R_{set} decreases [upper panel of Fig. 2(b)]. When $R_{set} < \sim 20\ \Omega$ (or $R_{filament} < 40\,000\ \Omega$), the top side lateral growth becomes saturated while the bottom side grows continuously, which means that the filament becomes more cylindrical. The estimated r range (3.5–5 nm) shown in Fig. 1(b) coincides very well with the experimentally observed diameter of the filaments [5–10 nm(r=2.5–5 nm)].[6]

We thank Professor Mi-Young Kim for the critical reading of the manuscript. This study was supported by the National Program for 0.1Terabit NVM Devices.

[1] B. J. Choi, D. S. Jeong, S. K. Kim, S. Choi, J. H. Oh, C. Rohde, H. J. Kim, C. S. Hwang, K. Szot, R. Waser, B. Reichenberg, and S. Tiedke, J. Appl. Phys. **98**, 033715 (2005).
[2] S. Seo, M. J. Lee, D. H. Seo, E. J. Jeoung, D.-S. Suh, Y. S. Joung, I. K. Yoo, I. R. Hwang, S. H. Kim, I. S. Byun, J.-S. Kim, J. S. Choi, and B. H. Park, Appl. Phys. Lett. **85**, 5655 (2004).
[3] S. C. Chae, J. S. Lee, S. Kim, S. B. Lee, S. H. Chang, C. Liu, B. Kahng, H. Shin, D.-W. Kim, C. U. Jung, S. Seo, M.-J. Lee, and T. W. Noh, Adv. Mater. (Weinheim, Ger.) **20**, 1154 (2008).
[4] S. B. Lee, S. C. Chae, S. H. Chang, J. S. Lee, S. Seo, B. Kahng, and T. W. Noh, Appl. Phys. Lett. **93**, 212105 (2008).
[5] K. M. Kim, B. J. Choi, Y. C. Shin, S. Choi, and C. S. Hwang, Appl. Phys. Lett. **91**, 012907 (2007).
[6] D. H. Kwon, K. M. Kim, C. S. Hwang, and M. Kim, Microstructure Analysis of Conducting Path in TiO_2 film presented at Fall meeting Mater. Res. Soc., Boston, MA, Dec. 1–5, 2008.
[7] K. M. Kim, B. J. Choi, B. W. Koo, S. Choi, D. S. Jeong, and C. S. Hwang, Electrochem. Solid-State Lett. **9**, G343 (2006).
[8] K. M. Kim, B. J. Choi, D. S. Jeong, C. S. Hwang, and S. Han, Appl. Phys. Lett. **89**, 162912 (2006).
[9] K. M. Kim, B. J. Choi, and C. S. Hwang, Appl. Phys. Lett. **90**, 242906 (2007).
[10] C. Rohde, B. J. Choi, D. S. Jeong, S. Choi, J.-S. Zhao, and C. S. Hwang, Appl. Phys. Lett. **86**, 262907 (2005).
[11] R. F. Bartholomew and D. R. Frankl, Phys. Rev. **187**, 828 (1969).
[12] U. Russo, D. Ielmini, C. Cagli, A. L. Lacaita, S. Spiga, C. Wiemer, M. Perego, and M. Fanciulli, Tech. Dig. - Int. Electron Devices Meet. **2007**, 775.