Influence of the Cu-Te composition and microstructure on the resistive switching of Cu-Te/Al2O3/Si cells — Source link ↗

Ludovic Goux, Karl Opsomer, Robin Degraeve, Robert Muller ...+5 more authors

Related papers:

- Redox-Based Resistive Switching Memories – Nanoionic Mechanisms, Prospects, and Challenges

- Nanoionics-based resistive switching memories

- Electrochemical metallization memories—fundamentals, applications, prospects

- Observation of conducting filament growth in nanoscale resistive memories

- Electrochemical dynamics of nanoscale metallic inclusions in dielectrics

Influence of the Cu-Te composition and microstructure on the resistive switching of Cu-Te/Al2O3/Si cells

L. Goux, K. Opsomer, R. Degraeve, R. Müller, C. Detavernier et al.

Related Articles

Kelvin probe force gradient microscopy of charge dissipation in nano thin dielectric layers
J. Appl. Phys. 110, 084304 (2011)

Kelvin probe study on formation of electric dipole at direct-contact HfO2/Si interfaces
J. Appl. Phys. 110, 074115 (2011)

Insight into unusual impurity absorbability of GeO2 in GeO2/Ge stacks
Appl. Phys. Lett. 99, 142101 (2011)

Electrical properties of atomic layer deposited aluminum oxide on gallium nitride
Appl. Phys. Lett. 99, 133503 (2011)

Characterization of channel strain evolution upon the silicidation of recessed source/drain Si1xGex structures
Appl. Phys. Lett. 99, 133107 (2011)

Additional information on Appl. Phys. Lett.

Journal Homepage: http://apl.aip.org/

Journal Information: http://apl.aip.org/about/about_the_journal

Top downloads: http://apl.aip.org/features/most_downloaded

Information for Authors: http://apl.aip.org/authors

Influence of the Cu-Te composition and microstructure on the resistive switching of Cu-Te/Al$_2$O$_3$/Si cells

L. Goux,[1,a)] K. Opsomer,[1] R. Degraeve,[1] R. Müller,[1] C. Detavernier,[2] D. J. Wouters,[1] M. Jurczak,[1] L. Altimime,[1] and J. A. Kittl[1]
[1]IMEC, Kapeldreef 75, 3001 Leuven, Belgium
[2]Universiteit Gent, Krijgslaan 281 (S1), 9000 Gent, Belgium

In this letter, we explore the influence of the Cu$_x$Te$_{1-x}$ layer composition ($0.2 < x < 0.8$) on the resistive switching of Cu$_x$Te$_{1-x}$/Al$_2$O$_3$/Si cells. While $x > 0.7$ leads to large reset power, similar to pure-Cu electrodes, $x < 0.3$ results in volatile forming properties. The intermediate range $0.5 < x < 0.7$ shows optimum memory properties, featuring improved control of filament programming using <5 μA as well as state stability at 85 °C. The composition-dependent programming control and filament stability are closely associated with the phases in the Cu$_x$Te$_{1-x}$ layer and are explained as related to the chemical affinity between Cu and Te. © *2011 American Institute of Physics*. [doi:10.1063/1.3621835]

Resistive Random Access Memory (RRAM) technology has drawn a lot of attention these last years due to scaling potential. Between the different RRAM concepts, the Conducting-Bridging RAM (CBRAM) was demonstrated to show attractive properties, such as endurance robustness and multi-level capability.[1,2] A CBRAM cell typically consists of a cation-supply electrode, usually based on Cu or Ag metal, an insulating layer serving as electrolyte for metal-cation drift, and an electrochemically inert electrode. A positive electrical potential applied to the former electrode results in the filament growth from the cathode towards the cation-supply electrode, bridging eventually the two electrodes and leading thus to the electrical switching to a low-resistive state (LRS). On the other hand, a negative potential induces the reverse redox reaction, i.e., the filament dissolution and cation drift back towards the cation-supply electrode, accounting in turn for the return to a high-resistive state (HRS).

While chalcogenide materials like GeSe or GeS were originally developed as electrolyte layers,[1,2] binary-oxide media like HfO$_2$,[3] ZrO$_2$,[4] or Ta$_2$O$_5$ (Ref. 5) were more recently introduced. Combined with a pure-Cu supply layer, oxide-based CBRAM cells typically exhibit attractive switching properties and filament stability, however they usually require larger reset current I_{RESET}[3–5] than chalcogenide-based CBRAM cells. On the other hand, the Cu-Te alloy has also recently been introduced as a Cu-supply layer.[6]

In this work, we explore the effect of the Cu$_x$Te$_{1-x}$ composition on the switching characteristics of an original and integration-friendly Cu$_x$Te$_{1-x}$/Al$_2$O$_3$/Si cell. We particularly show that the composition range $0.5 < x < 0.7$ results in controlled filament generation and optimized memory characteristics.

A 3 nm-thick Al$_2$O$_3$ layer was deposited on doped-Si wafers by an H$_2$O-based Atomic Layer Deposition (ALD) technique. Then, 3 mm-wide and 50 nm-thick Cu$_x$Te$_{1-x}$ dots were deposited through a shadow mask by a co-sputtering

process from a Cu target and a Cu$_{0.1}$Te$_{0.9}$ target. Finally, 1 mm-wide Pt dots were sputtered on top of the Cu$_x$Te$_{1-x}$ dots using a different shadow mask. Fig. 1(a) shows schematics of the cells.

A combinatorial approach was used to study the effect of Cu$_x$Te$_{1-x}$ composition. During the co-sputtering process of the Cu$_x$Te$_{1-x}$ layer, triangular masks placed in front of the sputtering targets result in a composition gradient in the deposited film, with a gradual variation of x along a direction of the wafer, called y-direction hereafter. Fig. 1(b) shows the composition of the Cu$_x$Te$_{1-x}$ layer determined by X-ray fluorescence (XRF) spectroscopy for each position along the y-direction. Using the 4-point probing method, we verified that the Cu$_x$Te$_{1-x}$ layer exhibits low resistivity in the whole composition range [Fig. 1(c)].

The Pt/Cu$_x$Te$_{1-x}$/Al$_2$O$_3$/Si cells were electrically tested using a conventional parameter analyzer HP4156. No load resistor was used. Current-Voltage (I-V) characterization was carried out by applying positive voltage to the Pt top-electrode (TE) for forming and set operations, and negative voltage for reset operation, in agreement with CBRAM switching mode. Note that no switching is obtained when forming is performed using negative voltage ramps.

Fig. 2(a) shows the initial resistance R_{INIT} of the as-prepared cells together with the forming voltage V_F as a function of x in Cu$_x$Te$_{1-x}$. On the one hand, R_{INIT} steadily decreases with the increase of x, suggesting enhanced Cu in-diffusion in the Al$_2$O$_3$ layer for larger x. On the other hand, V_F shows a strong drop for $x > 0.7$. The X-ray diffraction (XRD) patterns performed on the Cu$_x$Te$_{1-x}$ layers [Fig. 2(b)] clearly show different microstructures depending on the composition. The Cu$_x$Te$_{1-x}$ layers are amorphous for $x < 0.5$. XRD peaks corresponding to crystalline Cu-Te phases are identified for $x > 0.5$, while peaks corresponding to crystalline Cu become clearly visible only for $x > 0.7$. These results are in agreement with the Cu-Te phase diagram.[7–9] The composition range $x > 0.7$ also showed a marked difference of surface morphology compared to $0.5 < x < 0.7$. Large protrusions, presumably associated with
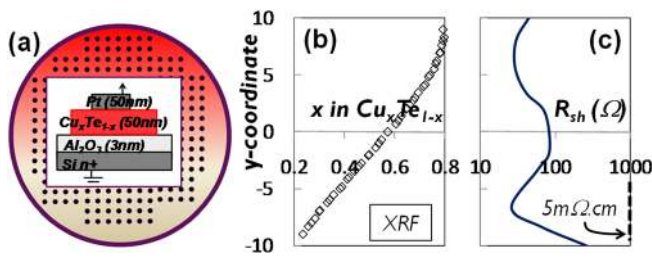
a)Electronic mail: gouxl@imec.be

FIG. 1. (Color online) (a) Cu_xTe_{1-x} dot pattern on the wafer, the inset showing a schematic cross-section of the cell stack; (b) and (c) relations between the y-coordinate of the cell and the composition and sheet resistance R_{sh}, respectively, of the Cu_xTe_{1-x} layer.

the presence of the pure Cu cubic phase, were indeed observed in the range x > 0.7 [Fig. 2(c)]. Hence, the drop of V_F for x > 0.7 may be associated to the presence of the Cu cubic phase in the Cu-Te layer. This result is in agreement with a recent report showing shorter time to dielectric breakdown for dielectrics having Cu gates instead of electrochemically inert Pt gates.[5]

The Cu_xTe_{1-x} composition also strongly affects subsequent resistive switching, in particular the filament characteristics. Figs. 3(a) and 3(b) show, respectively, the LRS resistance R_{LRS} and the reset current I_{RESET} extracted from negative voltage ramps performed after set switching operations using the compliance currents $I_{COMP} = 100$ μA or $I_{COMP} = 5$ μA. Larger I_{COMP} leads to lower R_{LRS} and larger I_{RESET}, as widely reported in the literature for RRAM (Ref. 10) and CBRAM (Refs. 11 and 12) devices. In addition, three main composition regions may be distinguished, associated with the typical I-V traces shown in Fig. 3(c). For region (1) x < 0.5, very unstable Cu filament is generated [Figs. 3(a) and 3(b)], and the lower x values typically result in volatile forming characteristics [Fig. 3(c)], which may
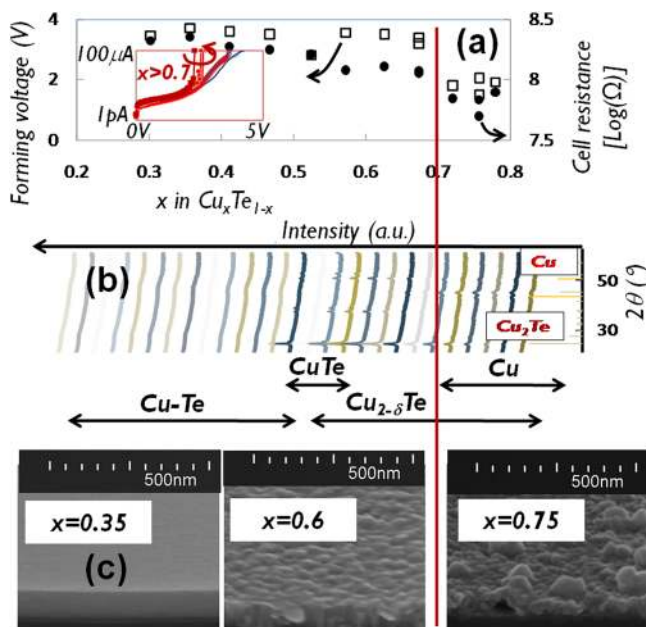

FIG. 2. (Color online) (a) Forming voltage V_F and resistance R_{INIT} of as-prepared cells (measured at +0.2V) as a function of x in Cu_xTe_{1-x}; the inset shows the forming I-V traces; (b) XRD diagrams of the Cu_xTe_{1-x} layers, revealing different phases depending on the x range; (c) Scanning Electron Microscopy (SEM) images showing the surface morphologies of the Cu_xTe_{1-x} layer for different x values.

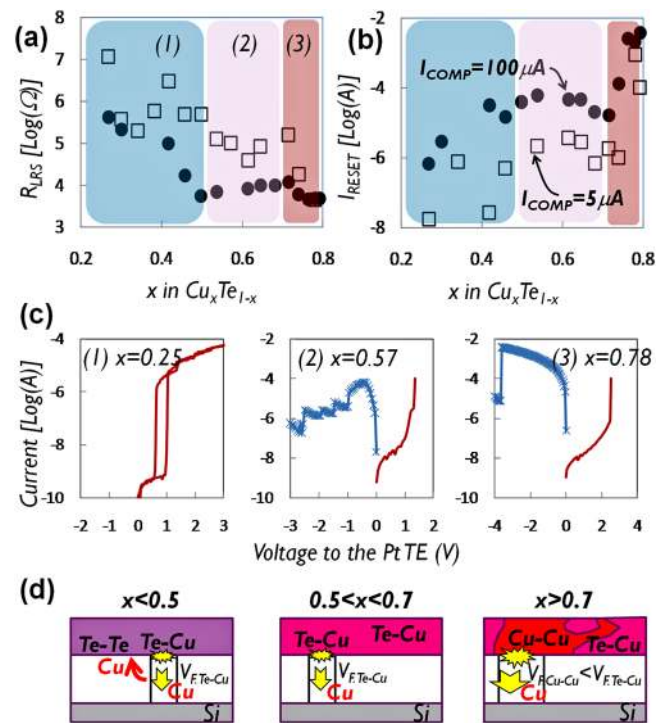FIG. 3. (Color online) LRS resistance R_{LRS} (a) and reset current I_{RESET} (b) as a function of x in Cu_xTe_{1-x}, both extracted after set switching using $I_{COMP} = 100$ μA (full circles) or $I_{COMP} = 5$ μA (empty squares); (c) Typical I-V set and reset traces observed for the three composition regions (1) x < 0.5, (2) 0.5 < x < 0.7, and (3) x > 0.7; (d) Schematics of the phenomenology of Cu-filament formation in each region.

appear attractive for diode-selector applications. For region (3) x > 0.7, a stable Cu filament is formed, however requiring large reset current $I_{RESET} \gg I_{COMP}$, similar to pure-Cu supply layers.[3–5] We also verified this effect by using pure-Cu dots instead of Cu-Te material (not shown here). I-V cycling tests performed in region (3) resulted in >10% switching failures, probably related to large switching power. On the other hand, region (2) 0.5 < x < 0.7 shows attractive switching control using low operation current $I_{RESET} \sim I_{COMP}$. Fig. 4(a) confirms the remarkable resistance stability observed in region (2) over 10^3 I-V cycles using <5 μA. Filament stability in region (2) is also assessed up to more than 10^4 s at 85 °C [Fig. 4(b)]. Similar retention was obtained in region (3).

Hence, the composition range 0.5 < x < 0.7 in Cu_xTe_{1-x} appears as the best trade-off in terms of programming control, limited operation current, and filament stability. The good control of low I_{RESET} by I_{COMP} suggests very little
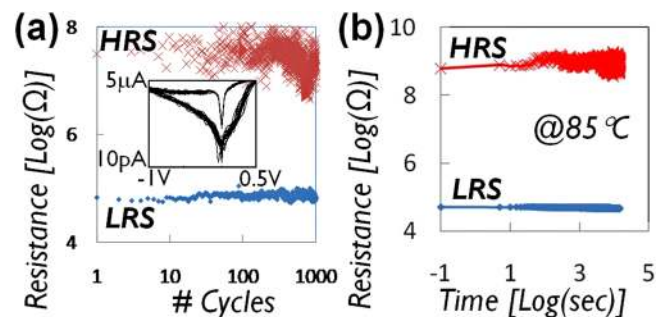

FIG. 4. (Color online) (a) HRS and LRS resistance control observed over 10^3 I-V cycles using $I_{COMP} = 5$ μA (inset shows 30 consecutive I-V cycles); (b) HRS and LRS resistance monitoring at 85 °C using constant bias voltage of 20 mV.

sensitivity to the parasitic current overshoot induced by forming/set.[10] We attribute this property to the larger energy barrier for Cu injection from Cu-Te crystal phases compared to pure Cu, due to larger Cu-Te bonding energy. In agreement, Da Silva *et al.* reported density-functional-theory calculations showing that in this composition range the formation energy of Cu-Te structures is lower than the formation energy of pure Cu phase.[7] Therefore, the Cu injection into the Al_2O_3 layer requires less energy from the Cu cubic phase, which results in a lower V_F values measured for $x > 0.7$ [see inset of Fig. 2(a)]. This also accounts for an increased sensitivity of the cell to discharge current in this range, leading to large Cu electromigration and resulting in overgrown filaments which will require larger I_{RESET}. On the other hand, for $x < 0.5$, the better stability of CuTe and Te phases[8,9] probably does not allow any stable Cu phase formation, so that the Cu atoms injected at forming will readily diffuse back to the supply layer after power switch off. To the contrary, the stability of Cu filaments for $0.5 < x < 0.7$ may be explained by the stability of Cu-deficient $Cu_{2-\delta}Te$ phases in this range [Fig. 2(b)]. Note that the decrease of R_{INIT} already observed in this range might also originate from enhanced mobility of Cu in this range.[7] Fig. 3(d) shows schematics of the filament formation scenario in each region.

To summarize, we showed the strong influence of x in the Cu_xTe_{1-x} layer on the memory properties of Cu-Te/Al_2O_3/Si cells, and we associated this influence to the different Cu-Te phase stability regions. Improved control of the filament formation is obtained in the range $0.5 < x < 0.7$ compared to $x > 0.7$, which we attribute to the Cu-Te bonding energy of the phases in the former range. The cell is programmed using < 5 μA current level without transistor-based current limiter, excellent stability of programming parameters is demonstrated over 10^3 cycles, and state stability was verified up to 10^4 s at $85\,^\circ$C.

[1]M. N. Kozicki, M. Park, and M. Mitkova, IEEE Trans. Nanotechnol. **2**, 331 (2005).

[2]M. Kund, G. Beitel, C.-U. Pinnow, T. Röhr, J. Schumann, R. Symanczyk, K.-D. Ufert, and G. Müller, Tech. Dig. - Int. Electron Devices Meet. **2005**, 754.

[3]Y. Wang, H. Lv, W. Wang, Q. Liu, S. Long, Q. Wang, Z. Huo, S. Zhang, Y. Li, Q. Zuo, W. Lian, J. Yang, and M. Liu, Electron Device Lett. **31**(12), 1470 (2010).

[4]Q. Liu, S. Long, W. Wang, Q. Zuo, S. Zhang, J. Chen, and M. Liu, Electron Device Lett. **30**(12), 1335 (2009).

[5]T. Tsuruoka, K. Terabe, T. Hasegawa, and M. Aono, Nanotechnology **21**, 425205 (2010).

[6]K. Aratani, K. Ohba, T. Mizuguchi, S. Yasuda, T. Shiimoto, T. Tsushima, T. Sone, K. Endo, A. Kouchiyama, S. Sasaki, A. Maesaka, N. Yamada, and H. Narisawa, Tech. Dig. - Int. Electron Devices Meet. **2005**, 783.

[7]J. L. F. Da Silva, S.-H. Wei, J. Zhou, and X. Wu, Appl. Phys. Lett. **91**, 091902 (2007).

[8]P. F. Carcia, F. D. Kalk, P. E. Bierstedt, A. Ferretti, G. A. Jones, and D. G. Swartzfager, J. Appl. Phys. **64**(4), 1671 (1988).

[9]S. K. Chattopadhyay, S. K. Chatterjeet, and S. P. Sen Gupta, J. Phys. D: Appl. Phys. **22**, 142 (1989).

[10]L. Goux, J. G. Lisoni, X.P. Wang, M. Jurczak, and D. J. Wouters, IEEE Trans. Electron Devices **56**(10), 2363 (2009).

[11]C. Schindler, S. C. P. Thermadam, R. Waser, and M. N. Kozicki, IEEE Trans. Electron Devices, **54**(10), 2762 (2007).

[12]U. Russo, D. Kamalanathan, D. Ielmini, A. L. Lacaita, and M. N. Kozicki, IEEE Trans. Electron Devices **56**(5), 1040 (2009).