Role of temperature in valence change memory devices

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ARTICLE INFO

ABSTRACT

The I-V curve of forming, switching between SET and RESET and memory are evaluated theoretically. The key features of the model are: a) drift diffusion of ions and of electrons, b) stoichiometry changes by exchange of oxygen with the ambient, c) temperature changes with current, d) Mott transitions in forming the filament and e) switching by changes in tunneling through a thin insulating layer. Forming, SET and RESET are induced by stoichiometry changes. Both voltage ramp and voltage pulses are considered. For pulses the interplay between pulse duration and pulse voltage is evaluated.

1. Introduction

Valence change resistive switching random access memory (RRAM) technology is widely explored for non-volatile memory as well as for memristor applications [1–4]. In RRAM devices based on metal oxides, it is believed that resistance changes are induced by redistribution of oxygen vacancies, which act as electron donors. Temperature variations play a major role in the physics of valence change RRAM devices, and can account for the many order of magnitude difference between writing and retention times. With increasing temperature, the mobility of the oxygen vacancies and the oxygen exchange coefficient of the electrodes rises exponentially and oxygen stoichiometry can readily be changed and oxygen vacancies be redistributed. After reaching a new distribution, by reducing the temperature, the new vacancy distribution is frozen-in. It is thus of major importance to include temperature related effects when modeling valence change devices. Menzel et al. [4] have considered the temperature dependence of the ion mobility in their simulations of RRAM devices by exponential thermal activation of the ions mobility. They ascribed switching to migration of oxygen vacancies through an insulating gap, but used an ad-hoc equation to depict the filament edge. Ielmini [5] also takes into consideration temperature, but investigates thermal radial gradient which affects the filament diameter. This behavior explains unipolar switching [6] and was explored theoretically by Kim et al. [3]. Our work focuses on bipolar switching.

Abrupt switching must be due to a physical mechanism which allows significant changes in the electrical resistance under minor changes in the oxygen vacancy distribution [7]. Tunneling through a narrow insulating gap without band bending or through a narrow Schottky barrier can thus account for switching. We consider tunneling, hence the model applies when a tunneling gap exists. When no tunneling gap exists, a Mott (insulator to metal) transition may account for the abrupt transition. While we assume that a conducting section (representing a filament in 3D) exists between the electrodes, due to a Mott transition, the abrupt switching behavior is due to tunneling, not due to the Mott transition.

While most of the past works focused only on one of several physical processes, lately work of Marchewka et al. [8–10] combines several physical processes. Tunneling takes place through a Schottky like potential barrier. The I-V curve for bipolar switching [8] exhibits on the first run, a clock wise time propagation. This does not provide an explanation for those experimental results reported so far, which exhibit counter clock wise propagation with time of the I-V curve during the first, electroforming run of bi-polar switching.

In this publication we incorporate temperature variations in the mixed ionic electronic transport equations of the bulk, which describe oxygen vacancies redistribution as well as in the oxygen exchange coefficient of the electrodes and the resulting changes in conductivity. Previously, these equations were solved only for the constant temperature case [11]. We consider a most simple 1-dimensional configuration, thus there is no bulk region surrounding the filament. In this model switching occurs due to modulation of an insulating gap between the electrode and the conducting section (filament). The gap modulation occurs by motion of ions at the tip of the filament. The gap size controls the tunneling of the electron. A one dimensional model is appropriate in case the motion of the ions, which are responsible to

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resistance changes, takes place predominately longitudinally, and heat transport takes place predominately towards the electrodes [9]. Our 1D model should capture the salient characteristics of devices exhibiting bi-polar switching. We model both electroforming and switching.

The aim of this paper is to demonstrate that a combination of five well known physical processes, but yet in 1D and simple set of equations, provides excellent qualitative modeling of the I-V relations both during forming and then switching. The processes are: a) drift diffusion of ions and of electrons, b) stoichiometry changes by exchange of oxygen with the ambient, c) heating by electrical power, d) forming the conducting section (filament) by a Mott transition and e) switching by tunneling through a thin insulating layer. The nature of the electrodes examined below results in a filament that grows from the anode side towards the cathode. The model allows calculation of retention time as well as endurance, and studying the interplay between pulse duration and pulse voltage. The model combines multiple major physical processes and still conserves it simplicity.

2. Model

The mixed-ionic-electronic-conductor (MIEC) modeled in this work includes two types of charge carriers: mobile donors and electrons. In many metal oxides oxygen vacancies are native donors, and can be doubly ionized. However, at low temperatures close to room temperature, the second ionization is expected to be quenched, as reported for several oxides near room temperature [12–14]. We assume here that donors are singly ionized or neutral throughout the entire temperature range considered. The ion charge considered is relative to the perfect crystal, according to Kröger-Vink notation. We further assume that oxygen, hence oxygen vacancies, can penetrate the top electrode but not the bottom electrode, due to the impervious substrate on which it is applied.

The pristine oxide is an MIEC with a relative high resistance for both electron and ion conduction. During forming, our model describes how oxygen vacancies (donors) exchanged through the top electrode with the environment, pile up and create a highly conducting metallic region. We assume that the metallic region is due to a Mott transition that takes place when the oxygen vacancy concentration exceeds a certain limit. The conducting region is formed from the anode side when the oxygen exchange rate at the top electrode is fast compared to the rate of oxygen vacancy redistribution in the oxide [9,15,16]. The remaining of the oxide between the tip of the metallic region and the cathode is the rather insulating MIEC responsible for the switching in our model, as schematically shown in Fig. 1. Through this gap two electron currents flow in parallel, a drift-diffusion one and a tunneling one. The latter becomes significant when the gap becomes small during electroforming. Small variations in the tunneling gap width result in large changes in the resistance of the sample. Tunneling is modeled by Fowler-Nordheim approximation [17,18],
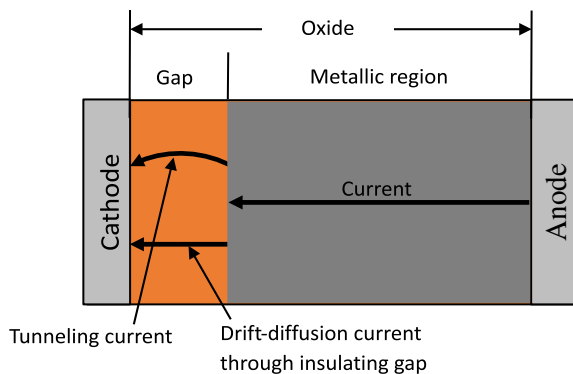

Fig. 1. Schematic current paths during electroforming and SET.

$$j_{e,tunneling} = AF^2 e^{-\frac{4\sqrt{2mq}\,\Phi_B^{3/2}}{3\hbar F}}, \quad A = \frac{q^2}{16\pi^2\hbar\Phi_B} \tag{1}$$

where Φ_B is the potential barrier between the insulator and electrode and $F \cong \Phi_B/L_{gap}$ is the electric field in the insulating gap, L_{gap} the gap width, q elementary charge and m the electron effective mass.

The temperature in the sample is assumed uniform in our model. This assumption is reasonable because Kim et al. [3] showed that the temperature is relatively uniform in the filament between 600 and 650 K. The temperature is calculated by balancing the input electrical power and heat dissipation through the electrodes $K_{th}(T - T_{RT})$ where K_{th} is the thermal boundary conductance. K_{th} which governs the heat dissipation from the oxide, depends significantly on the interface between the materials, and on temperature and we apply a T^{-3} temperature dependence of K_{th} [19,20].

Temperature plays a major role in our model. The mobility of the donors increases exponentially with temperature, due the thermally activated hopping from one site to a nearby one. We limit the donor mobility at high temperatures (because of the finite size of the potential barrier),

$$\nu_i^{-1} = \nu_{i,max}^{-1} + \nu_{i0}^{-1}e^{E_i/kT} \tag{2}$$

where k is the Boltzmann constant, and apply E_i the activation energy for ion hopping of 0.7–1 eV [21]. In the simulations we have assumed that $\nu_{i,max}/\nu_e(T = T_{RT}) = 10^3$, to stay in the regime in which the rate of redistribution of ions in the bulk is low compared to the rate of vacancy injection from the top electrode [15,16].

In the simulations we assume a linear drift-diffusion model for electrons and charged donors, as previously described in [11,22–24]. It is noted that in the insulting gap there is also an electron tunneling component to the current, Eq. (1). The drift-diffusion current density for electrons in the oxide, both in the insulating gap and the metallic region, is expressed as [25]:

$$j_e = \frac{\sigma_e}{q}\nabla\tilde{\mu}_e \tag{3}$$

where $\tilde{\mu}_e$ is the electron electrochemical potential, the conductivity is

$$\sigma_e = qn\nu_e \tag{4}$$

and n is the electron concentration. The electron mobility is,

$$\nu_e = \nu_{e0}e^{-E_e/kT} \tag{5}$$

where we choose in the simulations $\nu_e(T = T_{RT}) = 1$ cm^2/Vs and an activation energy $E_e = 0.05$ eV [3]. The mobility coefficient ν_{e0} in the insulating oxide is 6.9 cm^2/Vs. Where a Mott transition takes place ν_{e0} is increased. To allow a manageable simulation the increase is by a factor 10^3. A Mott transition occurs where the vacancy concentration exceeds a given limit. The limit chosen is 10^{21} cm^{-3}, a factor of $\times 10^2$ of the equilibrium concentration of 10^{19} cm^{-3}. The highly conductive region formed in this way represents a metallic filament.

The ion current density is,

$$j_i = -\frac{\sigma_i}{q}\nabla\tilde{\mu}_i \tag{6}$$

where $\tilde{\mu}_i$ is the ion electrochemical potential. The ion conductivity is,

$$\sigma_i = qN_D\nu_i \tag{7}$$

N_D is the donor concentration and q the elementary charge. The ion mobility is given by Eq. (2).

Denoting the electrical potential as φ,

$$\tilde{\mu}_e = \mu_e^0 + kT\ln n - q\varphi$$
$$\tilde{\mu}_i = \mu_i^0 + kT\ln N_D + q\varphi \tag{8}$$

where μ^0 is a standard chemical potential.

Eqs. (3) and (6) can be rewritten as,