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Voltage Tuning Effect on Metal-Insulator Phase Transitions in Vanadium Dioxide Thin Film

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Vanadium dioxide, VO_2 pulsed laser deposited layer on gold buffer substrate was electro-thermally actuated for the transition-voltage dependence study. The results present the transition temperature decrease to 37 °C with 1.5V applied voltage and affecting the slopes transition steepness leading to large values of the temperature coefficient resistance.This behavior was modeled and simulated using a finite element method (FEM) through Comsol Multiphysics software based on material properties extracted from experimental characterizations allowing for a more accurate numerical evaluation. The simulated electrical resistance function of applied voltage and temperature could be used to design a Vanadium dioxide based device and predict the operating mode. This work has a plethora of potential application such as a memristive device with $R_{ON}/R_{OFF}\approx10^2$ at 1V hysteresis width and biosensors operating at room temperature.

Keywords:vanadium dioxide; metal-insulator transition; voltage tuned metal-insulator transition; FEM modeling

I. INTRODUCTION

Vanadium dioxide (VO_2) is known as a thermally driven phase change material with transition from the (monoclinic M1) metallic phase to tetragonal (rutile R) insulating phase, that occurs at temperature approximately around 68°C (Shao Z *et al.*, 2018; Zhang *et al.*,2015; Park *et al.*, 2013). The most interesting aspect in VO_2 is the possibility to modify transition temperature values using external stimuli such as temperature changes, doping (Liang*et al.*, 2017), optical signals (Goncalves *et al.*, 2016) and electric field (Hao Rulong *et al.*, 2016). During the transition, abrupt changes occur in the electrical, optical, and mechanical properties of the material over several orders of magnitude. Vanadium dioxide (VO_2) has gained great interests for many innovative applications including smart windows (Fang Xu *et al.*, 2018), memristive data storage devices (Macaluso et al., 2014; Beaumont *et al.*, 2014 ; Jihoon Kim *et al.*, 2015), energy-efficient high-speed switches (You Zhou *et al.*, 2013 ; Joonseok *et al.*, 2014; Vitale*et al.*, 2016), ultra-steep transition slope transistors (Nikhil *et al.*, 2015; Vitale*et al.*, 2015), and gas sensors (Haitao *et al.*, 2012). Vanadium oxide is a promising material for biological and biomedical applications such as Cr-doped VO_2 as optically driven actuators with high deflections in fluids (Merced *et al.*, 2012) and VO_x as the biosensor to

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detects biological molecules in enzymatic reactions (Inomata *et al.*, 2016).

The difficulty for controlling the metal-insulator transition (MIT) temperature through an electrical signal is a main drawback for the development of integrated measuring systems (Shao Z *et al.*, 2018). It would be rather intriguing if the MIT-based temperature could be electrically tuned to set the sensor sensitivity and memristor characteristics. Hitherto, the foregoing paragraph aims to demonstrate the capability of an electrical signal to achieve for a high sensitive sensor, giving us the ability to adapt the operating point according to the working temperature. In this paper, we demonstrate experimental analysis which consists of electrical characterization for laser ablated vanadium dioxide thin film on gold (VO_2/Au).

The transition is thermally induced and tuned by applied voltage bias via two probes tips. To study the VO_2/Au thin-film electrical switching behavior, we present the simulation results using the finite element method (FEM). The simulation procedures are based on material properties extracted from experimental characterization to obtain accurate numerical modelling close to the actual behavior of the electrical resistance as a function of applied voltage sweeps. We investigate the voltage effects on transition characteristics of the VO_2/Au layer including; transition temperature value, transition slope sharpness and hysteresis behavior. Finally, based on the voltage tuning effect on the VO_2 transition, the potential applications in both electronics and biosensing are also discussed.

II. EXPERIMENTAL AND PROCEDURE

1. VO_2 thin film deposition

The structure in this study consists of a 500 nm VO_2 thin film deposited on 200 nm metallic gold Au (111) using laser-ablated vanadium dioxide thin film as illustrated in Figure 1. Detail for the experimental procedures are described in previous works (Hassein-Bey *et al.*, 2016; Lafane *et al.*, 2017). Previous work (Hassein-Bey *et al.*, 2017) detailed the cross-section morphology structure analysis of VO_2 thin film deposition using scanning electronic microscopy (SEM). This VO_2 crystal growthanalysis shows a good uniformity with an average grain size of 70-90 nm.

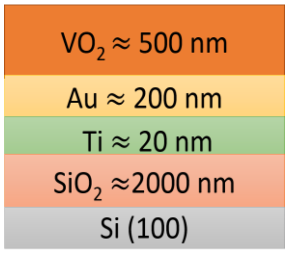

Figure 1. Schematic of the deposited VO_2 thin film

2. Experimental setup and characterization

We measure the current-voltage *(I–V)* characteristics of vanadium dioxide films in a wide temperature range from 35 to 100 °C with 5 °C step increment as shown in Figure 2. The top contact is performed by the deposition of micro-probe tips on the top surface of VO_2 thin film. A direct current (DC) characterization is carried out using Agilent HP4156C and Karl SussPA300 micromanipulator probe station. A hotplate is mounted inside Karl Suss AP4 micromanipulator probe station chuck to control the temperature. The polarization voltage range is varied between 0.03-1.5 V. Finally, the electrical resistance values R for each temperature T are deduced from the measured current-voltage characteristics *(R= R(T,V))*. The tuning effect of transition's temperature by applied voltage is of great interest in terms of ambient operating temperature and many unfolded potential applications could be developed. Practically in terms of operating temperature range many applications are targeted ranging from memristive to bio-sensing applications.

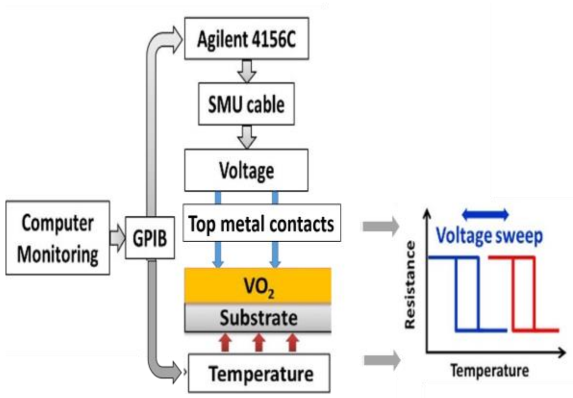

Figure 2. Experimental setup

3. Experimental Results and Discussions

The results for resistance characteristics are illustrated in Figure 3(a). This latter shows the temperature dependence of resistance for various voltages and demonstrates the transition temperature shifts to lower temperature around 37 °C with increasing voltage value. For instance, if the voltage bias value increases from 0.03 V to 1.5 V, the transition temperature values decrease from 70°C to 37 °C (see Figure 3(b)). For the greater voltage value, the transition slope increases gradually giving us a steeper transition's curves. The fact that transition temperature could be tuned by an external applied voltage to lower temperature, should be interpreted by a physical process of electrons injection in which electrons flow from the Au layer to the VO_2 layer due to the lower work function of the Au metal. Thus the local electronic density changes which support of the Mott–Hubbard phase transition model for VO_2 (Orlianges et al., 2012). The gold layer may have an important role in making Mott's transition to be more dominant with weak voltage for lower temperatures compared to classical transition SPT at around 68°C.

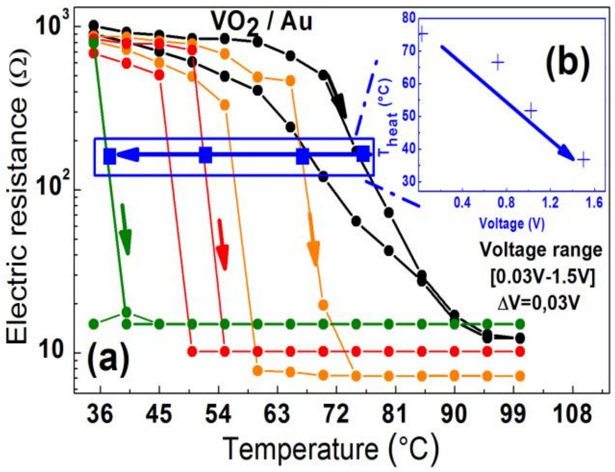

Figure 3. Thermally activated metal-insulator transition behavior in dependence with applied bias voltage for a metallic substrate (VO_2/ Au)

The steep zones in electrical resistance as a function of temperature are the zones of the high sensitivity of electrical resistance for a small variation in temperature (Figure 3). In general, these steep zones are localized around a fixed value of temperature i.e., MIT temperature (Shao Z et al., 2018). The capabilities to control the position of this latter zone and to shift it according to the working temperature can open a very interesting application field for high sensitivity thermal sensors and a more efficient memristor (Hassein-Bey *et al.*, 2016). In addition, it is very interesting to be able to tune the sensitivity value through a voltage signal and Figure 3 illustrates this possibility for some electrical polarization values. By shifting the polarization value from 0.03 V to 1.5 V, the transition zone moves from 76 ° C to 37 ° C .

III. MODELING AND SIMULATION OF TRANSITION OF VO_2 THIN FILM STRUCTURE

Modeling and simulation study play an important role in design and predict the device behaviour by giving the possibility to select the main design parameters for the sensor. The key- parameters to be controlled through our simulation are : (i) the temperature to achieve a high sensitivity value, (ii) and its mobility under the voltage bias. This simulation can develop devices based on the electric polarization that controls the thermal sensor sensitivity or the transition slope for the memristor device. Figure 3 allows us to extract the intrinsic thermoelectric properties of the materials and integrate them into the finite element simulation model as shown in Figure 4. This is to be the closest to the experimental reality in our design of sensors.

1. Modelling of thin-film structure

Figure 4 shows the proposed structure used in simulation which consists of two thin layers of 500 nm vanadium dioxide, VO_2, and 200 nm gold, Au. In order to study the electrical behaviour of the MIT material with voltage polarization and temperature, numerical simulations were performed using a finite element method (FEM).

The resistive response was modeled as a function of temperature and polarization voltage based on a two-point probes method. The two points probe has a spacing of approximately 4000 µm as in the experimental conditions (Hassein-Bey *et al.*, 2016). Thus a constant current is applied to the input of one probe and recovered from the second output. The resulting voltage between the two probes is calculated using the electric currents interface for conducting medium-thin layers in Comsol Multiphysics and the computed potential distributions (electric field).

By combining both the electrical voltage and the constant current into the two surface probes, we can deduce the material surface resistance.

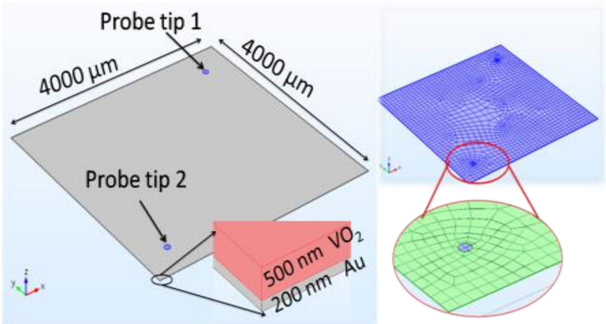

Figure 4. Proposed structure for simulation: The top layer is set to be VO_2, the bottom layer is set to have crystalline gold Au (111)

The modeling of the electric current flow in a conductive medium in the stationary case is governed by the continuity equation as:

$$\nabla\left(\mathbf{J}-\mathbf{J_e}+\frac{\partial \mathbf{D}}{\partial t}\right)=Q_i \quad (1)$$

Where **J** is the induced current density vector, **Je** is the external current density vector, **D** is the electric displacement field vector and Qi is the source current term. In general, Qi and **Je** are supposed to be null in our material. In the stationary mode, we could link the expression of the current density to the electric field **E** using the basic equation:

$$J=\sigma E \quad (2)$$

Where σ is the electrical conductivity. Thus, the electric resistance currents interface from Comsol Multiphysics is used to simulate the electric field, current, and potential distributions in the VO_2 thin layers. Based on Ohm's law, the software helps to solve a current conservation equation using the scalar electric potential as the dependent variable. The structure is meshed with a fixed layer swept mesh and in order to obtain the closest model to the actual behavior, the parameters describing the electrical material properties are directly extracted from experimental characterization results.

The main parameters in our model for the upper layer (VO_2) and bottom layer (Au) are given in Table 1.

Table 1. Main simulation parameters

Parameters	**Materials**	
	VO₂	***Au***
Resistivity	10^{+2} (Ω.cm)	4-15 (µΩ.cm)
Layer thickness	500 (nm)	200 (nm)
Lateral dimension	4000-6000 (µm)	4000-6000 (µm)

2. Simulation Results

This section describes the outcomes of the simulations to estimate the electrical behavior of the VO_2/Au thin layer for various temperatures, T and voltage V as illustrated in Figure 5. The curves in Figure 5 show a similar metal-insulator transition behavior with respect to polarization voltage bias. In fact, the two key-parameters are summarizing the variation of electrical resistance as a function of temperature: (i) Transition slope value and (ii) Transition temperature value. Based on the importance of a room temperature transition for targeted applications, we can predict by simulation the voltage polarization equivalent to 4.25 V.

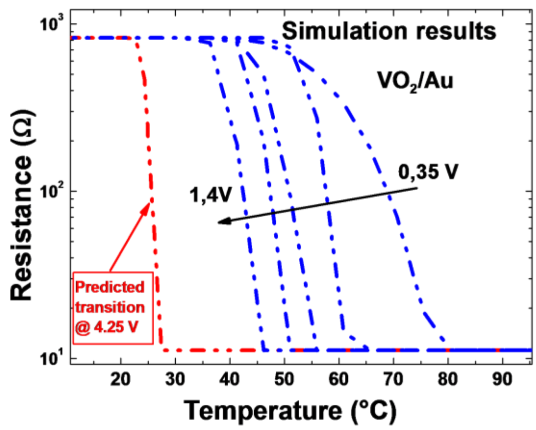

Figure 5. Change in electrical resistance R as a function of the temperature T for different polarization voltages by simulation

IV. POTENTIAL APPLICATIONS

In section II.3, we introduced experimental results and discussions for thermally activated metal-insulator transition behavior in dependence with applied bias voltage for a metallic substrate (VO_2/ Au). We show the ability to control and to shift the transition temperature with great sensitivity according to the desired working temperature. In this section, we present two potential applications based on the voltage tuning effect on metal-insulator phase transitions. We concern about the memory performance in VO_2/Au and the temperature sensing applications.

1. Memristive performance

The impending end of Moore's Law demands an innovative way to design a different logic devices and computational metaphysics and new ways must be explored to prepare for the post-silicon century challenge (Takasu et al., 2016). Memristors have been considered as one of the most promising and attractive devices for next-generation memory technology (Joshua Yang et al., 2008). It's advantages depends on the following features and performances: Non-volatility, smaller, faster, switch less than nanoseconds, power-efficient and super dense (Joshua Yang et al., 2008). Memory effects in transition metal oxides (TMO)'s have changed the research paradigm to the new field studies including TiO2 (Fryauf et al., 2015), MgO (Jimin Wang et al., 2014), VO_2 (Horacio Coy et al., 2014 ; Pellegrino et al., 2012 ; Jihoon Kim et al., 2015). VO_2 is one of the most notable materials due to its fast response time and a large range of accessible resistance values through a metal-to-insulator transition (MIT) (Macaluso et al., 2014; Sung-Hwan Bae et al., 2013-; Wilkie Olin-Ammentorp& SantoshKurinec, 2015).

We study and evaluate (I-V) characteristics of the VO_2/metal sample at a temperature of 35° C and presents the results in Figure 6. The hysteresis showing the width of about 1V. In addition, the triggering voltages Vup and Vdown are relatively low and respectively equal to 1.5V and 0.5V. Since the properties of the device are dependent on the memristors' previous memory state, the I-V measurement procedure is an important step to find the memristor characteristics.

Our structure was set to voltage biased from -3 V to 3 V and back to -3 V again. The setting was performed with 1 seconde-long steps of 30 mV.

Figure 7 shows a prominent memristive behavior describing a linear characteristic around zero with clockwise pinched hysteresis and a R_{ON}/R_{OFF} ratio approximately around 10^2. Figure 8 suggests that VO_2 (500 nm) /Au has a better R_{ON}/R_{OFF} ratio compared to the study reported by (Macaluso *et al.*, 2014) by using VO_2 (10 nm)/FTO/glass coated glass. Eventhough the thickness of our VO_2 is 500 nm, the structure demonstrate 10^2 in term of R_{ON}/R_{OFF} ratio. Macaluso et al.,in this case, achieve a value of R_{ON}/R_{OFF} ratio less than 10 for a thickness around 10 nm of VO_2. In the future, the work can be further improved to achieve a better R_{ON}/R_{OFF} ratio with lower VO_2 thicknesses.

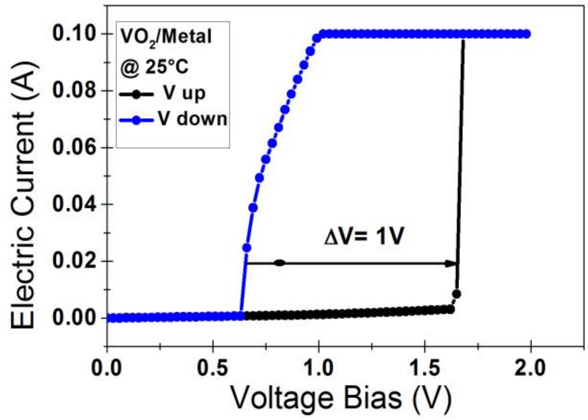

Figure 6. (I-V) characteristic of (VO_2/ Au) showing hysteresis width

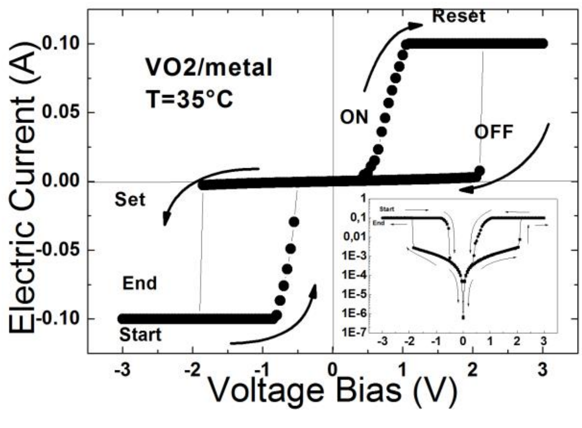

Figure 7. Characteristics of (VO_2/Au) samples showing a memristive behavior

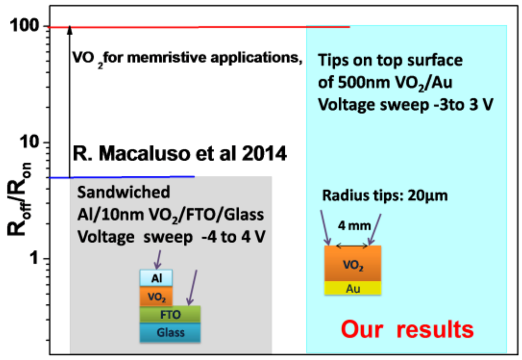

Figure 8. The R_{OFF}/R_{ON} ratio for 500nm VO_2/Au and 10nm VO_2/FTO

2. Biosensing performance

Based on the properties of VO_2, many potential applications could be developed especially for thermal biosensing technologies. The integrated vanadium dioxide in microfluidics device to detect the thermal activities in the biological process has good benefits for biosensors application (Li Wang *et al.*, 2008; Gurung *et al.*, 2013; Inomata *et al.*, 2016). In a thermal biosensor device, when a biological sample solution contains certain biochemical molecules, an interaction occurs with the sensitive layer and generates heat from the enzymatic reaction as shown in Figure 9. This allows us to detect a different type of disease by means of specific heat emission from their biological response. (Parikha Mehrotra, 2016; Inomata *et al.*, 2016). The working principle of the transducer element for thermal biosensor is to convert a biological response into an electrical signal but in general, it has limited sensitivity. The pursue the new materials, however, can mitigate the limitation issue and improve the sensitivity of biosensor in the modern healthcare equipment such as microfluidic system (Lab on Chip, µTAS). Besides its benefits that require a minimal sample from the patient, the device could help the medical practitioner to make a quick and premature detection of disease. (Naoki Inomata *et al.*,) study a microfluidic device to investigate the performance of a VO_x thermistor-based calorimeter for glucose and cholesterol detections due to its large TCR properties for highly sensitive thermal sensing (Inomata *et al.*, 2016).

The temperature coefficient of resistance (TCR) is directly proportional to the electrical resistance sensitivity (ΔR/R) and the temperature variation (ΔT) and is given by:

$$\Delta R/R = TCR \, {}^{1}/_{\Delta R} \qquad (3)$$

We are interested to figure out the effect of voltage sweeping on the quality of TCR. For this reason, the TCR versus T curves of VO_2/Au are calculated from the R versus T curves for different value of the applied voltage as depicted in Figure 10. We found that the gold layer has a considerable effect to improve the sensitivity response of the sensor i.e., TCR. To estimate the sensitivity of material for highly sensitive thermal sensing, usually, TCR is calculated relative to a thermal sweep (Equ. 3). However, in our study, we add a new voltage bias parameter to focus on voltage TCR's tuning effect.

For low voltage value, the TCR curve reflects a broad transition as low as 14.43 %/°C for a 0.03 V (see black color curve in Figure 10). For instance, at 0.51 V (refer red color curve in Figure 10), the TCR value increase to 52.36 %/°C at 63.9°C and TCR reach to 75.02 %/°C at 49.3 °C for a bias value of 1.02 V (see purple color curve in Figure 10). Simply put, as the operating voltage bias increases, the TCR has greater value and the size of the windows become narrower. Thus a careful choice for the operating point is very important in biosensor design. Basically, we have to select the higher TCR value but to obtain a more stable biosensor device and avoid noise problem, it is necessary to place the working point not too close to this zone. For instance, If the operating point is positioned around zone B (see Figure 10), the TCR will be approximately at 37 %/°C which is useful for highly sensitive thermal sensing application.

Based on the work of (Inomata *et al.*, 2016), the TCR value recorded 1.34%/°C for vanadium oxide (VO_x) has potential development for detecting biological molecules based on enzymatic reactions. Our result predicts a better and more interesting TCR value at 37 %/°C for a VO_2 that directly affected by a voltage bias. It provides more sensitivity to measure the generated heat from exothermic enzymatic reactions. This could encourage the development of biosensors based on enzymatic reactions for biological applications.

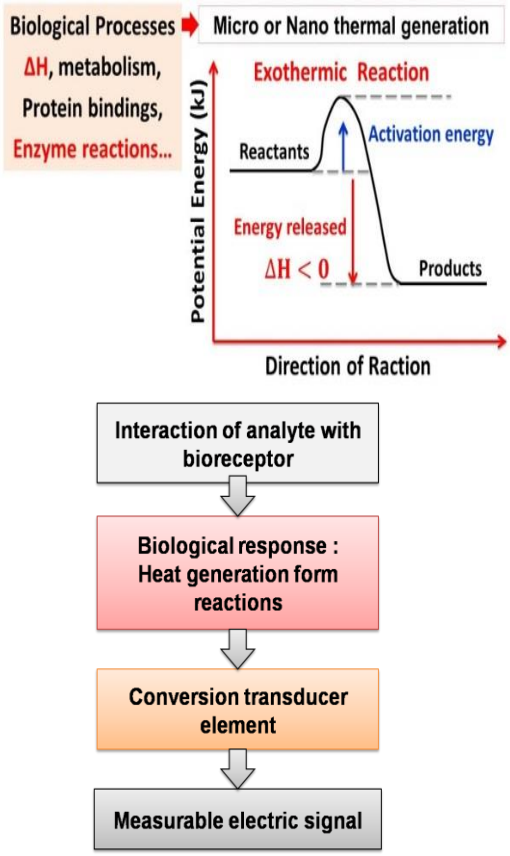

Figure 9. A diagram explaining the working principle of thermal biosensor and exothermic reactions occurring in biological processes

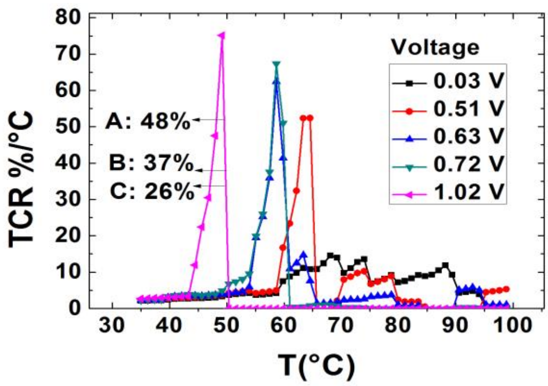

Figure 10. TCR curves for differents voltage values

V. CONCLUSION

Vanadium dioxide, VO_2 pulsed laser deposited layer on gold buffer substrate was electro-thermally actuated for the transition-voltage dependence study. In this paper, we demonstrate experimental analysis which consists of the electrical characterization of vanadium dioxide thin film on gold (VO_2/Au) deposited by the laser ablation technique. The transition is thermally induced and tuned by applied voltage bias via two probes tips. In addition, we present a finite element method (FEM) simulations to study the VO_2/Au thin-film electrical switching behavior. Results demonstrate the transition temperature shifts to a lower temperature around 37 °C with increasing voltage value. In addition, for greater voltage value, the transition slope gradually increases giving us a steeper transition's curves. Our numerical model restores a similar temperature dependence behavior of the electrical resistance with polarization voltage and predicts a transition at room-temperature under a voltage of 4.25 V.

Our sample presents a prominent memristive behavior describing a linear characteristic around zero with clockwise pinched hysteresis and a R_{ON}/R_{OFF} ratio approximately around 10^2. Eventhough the thickness of our VO_2 is 500 nm, the structure demonstrate 10^2 in term of R_{ON}/R_{OFF} ratio. In the future, the work can be further improved to achieve a better R_{ON}/R_{OFF} ratio with lower VO_2 thicknesses. Our results predict a better TCR value around 37 %/°C for a VO_2 to provide more sensitive biosensors based on enzymatic reactions. The finding could help to design a highly sensitive biosensor and open a new horizon for developing similar smart materials based on ambient temperature transition in the future. We show the ability to control and to shift the transition temperature with great sensitivity according to the desired working temperature. This works could be used as a powerful tool for designing several MIT-based sensors and predict the feasibility for a variety of devices with a high sensitivity of ambient temperature. Practically, in terms of operating temperature range, many applications are targeted going from the data storage devices to biosensors.

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