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MemSens: Memristor-Based Radiation Sensor

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***Abstract*— Resistive Random-Access memory (RRAM) technology has been gaining importance due to scalability, low power, non-volatility, and the ability to perform in-memory computing. The RRAM sensing applications have also emerged to enable single RRAM technology platforms which include sensing, data storage, and computing. This paper reports on sol-gel drop coated low-power μ-thick Ag/TiO_2/Cu memristor, named MemSens, developed for radiation sensing. MemSens exhibits a bipolar memristive switching behavior within a small voltage window, ranging up to +0.7 V for the turn-ON, and down to -0.2 V for the turn-OFF. Under these operating conditions, MemSens has 67% less switching voltage, 20% drop in ON switching current, 75% reduced active area and > 3x improved device endurance, compared to the best characteristics reported in the literature for μ-thick memristors. The device is tested under direct exposure to ionizing Cs-137 662keV γ-rays, during which a significant increase in the electrical conductivity of the device is observed. MemSens circuit is proposed to allow a relatively real time and cost-effective radiation detection. This provides a first insight to the advancement of reliable memristors that could potentially be deployed in future low-power radiation sensing technologies for medical, personal protection and other field applications.**

***Index Terms*— low power, sol-gel, endurance, active, bipolar, radiation, crossbar**

I. Introduction

MEMRISTORS, the fourth fundamental circuit element, have fulfilled the missing link between magnetic flux and electric charge in the same way resistor connects voltage to current, capacitor links voltage to charge and inductors link flux to current. Several nano-scale devices that have the same characteristics as memristors were observed before and after

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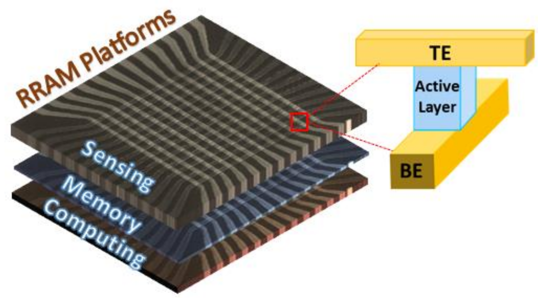

Fig. 1. System-on-RRAM for sensing, memory and computing.

Chua's postulation [1]. However, the connection to Chua's fundamental passive device was realized when HP Labs demonstrated the existence of a physical memristor [2]. This discovery sparked great interest in memristive devices and systems as one of the possible future alternatives to current CMOS-based memory technology for significant improvement in scalability, power consumption and performance [3]. In addition to supporting the traditional memory paradigm, considerable attention is focused on memristive device utilization for intelligent computing. Thus, memristor technology holds great potential to play a role in achieving close to ideal solution by enabling In-Memory-Computing (IMC) for both digital- and analog-type operations. This means computation and data storage can be accomplished at the same cell. Beside reducing the memory bottleneck, IMC makes it possible for energy-constrained systems to do both local processing and decision making, enabling autonomous operation [3]-[7].

Furthermore, memristor fingerprint *I-V* characteristic may alter against specific environmental changes, which generates the interest toward device deployment in sensing applications [8]-[14]. As shown in Fig. 1, advancements in memristor-based sensing will pave the way for system-on-RRAM platforms, where sensing, data storage, and computing are performed on the same memristor technology. This could significantly improve the overall cost, power, area and speed of the system.

Even though the field of memristive-based sensing is yet to be investigated, biological applications gain the highest importance, where memristor sensors can significantly reduce the overall power and cost of the system [14], [15]. On the other hand, few studies have been conducted to specifically investigate the radiation detection capability of memristor

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devices. These studies reported the radiation hardiness of nano-thick memristors, as the ultra-thin film acts as transparent layer under radiation exposure [16]-[22]. The work proposed in [23] by our research group was the first initial result to experimentally show γ-ray detection ability in a μ-thick TiO_2 based memristor device. Under the application of the non-switching bias voltage of 0.5 V, the structure proposed in [23] showed switching ability when exposed to gamma radiation.

This paper presents a μ-thick memristor Ag/TiO_2/Cu device fabricated by sol-gel drop-coating and denoted as MemSens. Compared to the memristor devices documented in [23], MemSens demonstrates:

- 67% reduction in the switching voltage.
- 20% drop in the ON switching current.
- 75% reduced active area.
- > 3x improved device endurance.

Moreover, a MemSens circuit is proposed and tested to allow relatively real-time and low power active radiation detection, under the bias voltage of 0.15 V. To the best of our knowledge, this is the first work that presents a low power and high-density radiation detection approach using memristor devices, which can smooth the path towards the advancement in system-on-RRAM platforms to perform sensing, data storage, and computing on the same memristor technology.

The body of the paper is organized as follows. Section II presents the fabrication process and the characterization methodologies used for the devices. The switching behavior of the fabricated memristor is explained in Section III. Section IV discusses the radiation interaction and active detection mechanism in the device. MemSens circuit is detailed in section V, followed by conclusions in section VI.

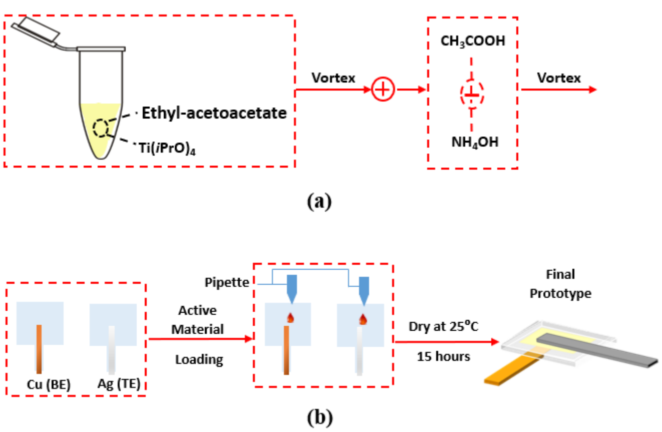

Fig. 2. (a) Active material (TiO_2) precursor solution synthesis. (b) Assembly of TiO_2 drop-coated orthogonal microscale memristor devices.

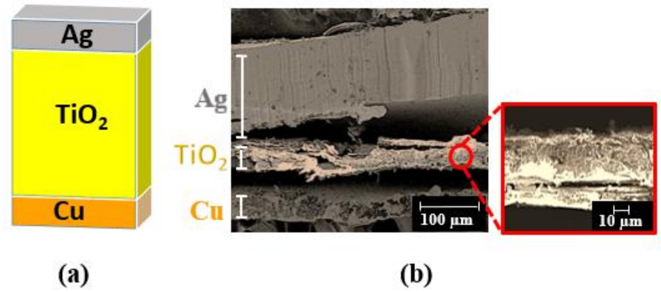

Fig. 3. (a) Cross-sectional representation of the MemSens device. (b) SEM photograph of a physical Ag/TiO_2/Cu memristor device, imaged in a cross-sectional view under secondary electron mode (accelerating voltage, 5 kV; magnification, x60; working distance, 12 mm).

II. Experimental Procedure

An iterative process of MemSens fabrication and characterization has been extensively carried out to achieve suitable physical and electrical characteristics desired for the radiation detection application.

A. *MemSens Fabrication*

The TiO_2 drop-coated memristors are fabricated by adjusting the sol-gel process described in [32]. In brief, ethyl-acetoacetate (99.9%) is added to titanium(IV) iso-propoxide, Ti(*i*PrO)$_4$, (98%) acting as a titanium source. Then, the system is vigorously mixed to obtain a homogenous yellowish transparent solution under room temperature conditions. Subsequently, ammonium acetate (99.6%) is added to the solution as a gelation agent. To construct the μ-thick memristor device, the electrode sheets, Ag and Cu, are mounted onto plastic substrates (Fig. 2 a,b). Then, the precursor solution is drop-coated on both electrodes. The coated material is left to dry at room temperature for fifteen hours. The device is finally stacked in a crossbar Ag/TiO_2/Cu structure, making a device area of 2 mm x 2 mm with an active layer thickness of approximately 45 μm (Fig. 3).

B. *MemSens Characterization*

Scanning electron microscopy imaging of MemSens is carried out using JSM-7610F Schottky field-emission microscope (JEOL LTD., Japan). Fig 3 (b) presents cross sectional morphology of MemSens. The thicknesses of Ag and Cu electrodes are 125 μm and 40 μm, respectively. The μ-thick TiO_2 layer is 45 μm in thickness with a dense and nonuniform geometry.

A Keithley 4200-SCS Parameter Analyzer (Tektronix) is used in the electrical measurement of the *I-V* characteristics of MemSens, using a dual voltage sweep mode. The prepared memristors are mounted on a probe station and are electrically tested by applying +1.4 V and -0.7 V onto the top electrode during the Set and Reset cycles, respectively. The compliance current (cc) is set to 200 μA during the Set process; whereas it

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is increased to the highest instrumental value (100 mA) during the Reset operation. A voltage pulse mode is used for holding the desired bias (0.15 V) during the irradiation studies.

III. MemSens Electrical Characteristics

A batch of MemSens, synthesized as explained in Section II, are assessed to demonstrate the *I-V* curve characteristics of the proposed device. An extensive characterization procedure is followed to investigate the optimal working conditions of the devices. For instance, gradually increasing voltage sweeps with different compliance current levels are used to identify low power, high resistance ratio, and high endurance device characteristics. This is accomplished under the application of +1.4 V and -0.7 V voltage sweeps to switch the device ON and OFF, respectively. As shown in Fig. 4, MemSens demonstrates a pinched hysteresis *I-V* curve with anti-clockwise and clockwise loops in the positive and negative voltage polarities, respectively. The obtained hysteresis loop confirms the expected electrical behavior for memristor devices [1], [2]. As illustrated in Fig. 4, MemSens exhibits repeatable *I-V* electrical profiles, and a relatively improved endurance (>50 consecutive hysteresis) compared with microscale devices, based on Al/TiO_2/Al and Al/TiO_2/Cu structures [23]. It can be observed that MemSens sets and resets in a classic bipolar behavior, yet the sharp turn-OFF behavior further indicates synergistic electric field and Joule heating effects [24]. The switching operation at less than 0.7 V makes MemSens ideal for low power sensing applications. Generally, memristor operation has tradeoff between the applied voltage, the switching time and the obtained resistance ratio. Thus, these parameters can be decided based on the desired applications. During MemSens characterization, the compliance current is set to 200 μA to reach a compromise between the device power, retention time and resistance ratio. Inset Fig. 4 shows the retention test results of MemSens. A voltage bias of 0.1 V amplitude and 0.05 s pulse width is applied every 15 minutes for 12 hours continuously, at room temperature. Shown inset illustrates that MemSens presented in this work has stable R_{OFF}/R_{ON} ratio of about 10^7. The stability in the resistance states is one of the electrochemical metallization mechanism (ECM) based-memristor features [24]. This is due to the robust filaments created during device Set and fully switched-OFF state obtained during Reset. The relatively long retention time and high resistance ratio further make MemSens ideal for sensing applications [23, 24].

The main target of this work is to utilize the switching ability of MemSens in radiation sensing. Therefore, it is of significant importance to examine the device behavior under DC voltage conditions. The aim is to investigate the optimal voltage bias that provides the needed junction for radiation interaction to take place in the device, while minimizing the synergistic effect of the electric field on the switching operation. To achieve this, MemSens is tested under the application of consecutive voltage pulses with different amplitudes (i.e. 0.1 – 0.5 V). The width of each pulse is equal to the switching time of the memristor associated to the applied voltage. For each voltage DC pulse, when device Set is achieved a Reset operation is performed to put the device back to its initial OFF state. As illustrated in Fig. 5, the ON switching time (when the device current increases from nA to μA range) substantially drops when the applied voltage increases from 0.1 V to 0.15 V, and it almost saturates for voltage pulses > 0.25 V. Various factors affect the speed of Set process such as the dissolution of the active electrode, the cation migration, the reduction of the ions to metal, and the filament growth [24]. Inset Fig. 5 reflects the exponential dependence of the device switching time on the applied electric field. To use the fabricated memristor as an active sensor, the optimal DC voltage value should be chosen to allow for low power operation while achieving a time window sufficient for radiation detection. A bias voltage of 0.15 which corresponds to the high gain region of the voltage-time plot (Fig. 5) has been selected for the radiation tests presented in the rest of the paper.

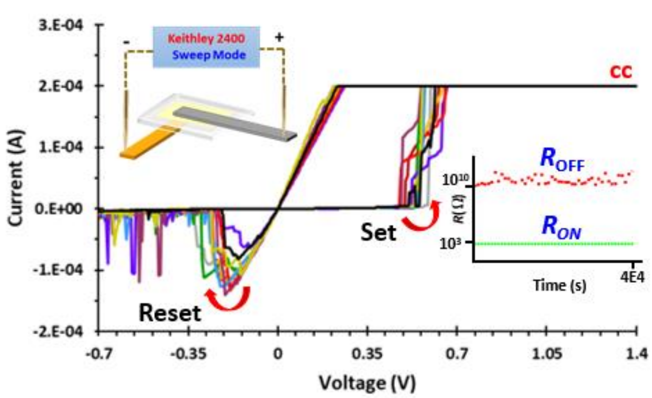

Fig. 4. Measured electrical characteristics of MemSens. The obtained *I-V* curve confirms the expected memristor behavior. cc, compliance current. Inset is the retention test results of MemSens.

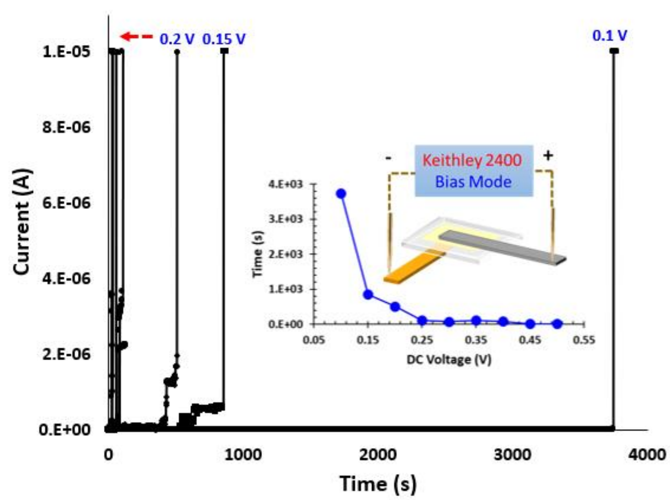

Fig. 5. Switching time recorded for different applied DC voltage pulses on MemSens.

IV. MemSens Radiation Detection Ability

The extensive electrical testing mentioned in Section III has enabled the use of MemSens for radiation detection experiments. As the electrical behavior of MemSens is well understood, especially under the application of DC voltage pulses, changes in device behavior under radiation exposure will be clearly detected. A Cesium-(Cs) 137 γ-ray emitting source (type-D disk, Eckert & Ziegler, Germany), with an active diameter of 5 mm, a radioactivity of 18.1 mCi (0.67 MBq), and a primary emitted photon energy of 662 keV is used during the radiation sensing experiments. Radiation exposure tests are performed by placing the radioactive source directly underneath MemSens, facing the silver electrode (inset Fig. 6). Radiation effects are monitored at room temperature, via real-time current measurement. Prior to radiation exposure, a fresh device is tested under the application of +1.4 V and -0.7 V to obtain the fingerprint *I-V* characteristic presented in Fig. 4, and consequently confirm the functionality of the device. Before testing MemSens in the active mode, passive exposure to a Cs-137 source is first carried out. After one day, radiation exposure is stopped and the device resistance is measured and compared to what is recorded before radiation exposure. This test confirms that MemSens is radiation hardened under passive exposure during OFF state. This property is utilized to propose crossbar based real-time monitoring, as explained in section V.

Based on the detailed study outlined in Section III, a bias voltage of 0.15 V is chosen to test MemSens for active sensing mode. This DC voltage is found to provide low power detection while maintaining high gain for the device switching times, with and without radiation. As shown in Fig. 6, a bias voltage of 0.15 V is applied until a turn-ON event is detected, then MemSens is reset and the DC test is repeated 12 times. It is clear that the minimum achieved switching time is about 360 s, in the absence of radiation. The same test is repeated after placing MemSens on Cs-137 source. The average ON switching time with radiation exposure has dropped by approximately 80%, compared to device behavior in the absence of radiation. However, the switching ability in the device is preserved, which serves the concept of utilizing the memristor crossbar for memory, sensing and computing.

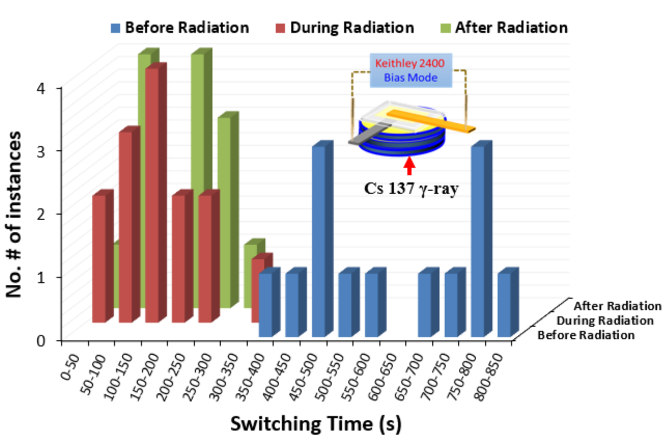

Fig. 6. Cs-137 radiation effect on the switching time of the fabricated $Ag/TiO_2/Cu$ devices under the application of 0.15 V clearly shows shorter switching time in the presence of radiation.

The dominant radiation interaction in MemSens is Compton scattering [25], [26], where a high energy photon collides with an electron, resulting in a recoil electron and a lower energy photon. While nearly independent of atomic number, Compton scattering probability is proportional to electron density in the material, and so is more likely to occur in the electrode, with events at the electrode-TiO_2 interface being most likely events for producing amplifiable signals. For an incident photon energy of 662 keV, recoil electrons have a continuous energy spectrum from ~180 keV (corresponding to the Compton edge) to ~490 keV (corresponding to the Compton backscatter peak). The mean free path of recoil electrons in TiO_2 consequently ranges from 0.18 μm to 0.43 μm, respectively [27]. In a simple, 1D calculation, recoil electrons from Compton backscattering can maximally impart ~60-70 eV to an oxygen atom during collisions with TiO_2 molecules. Thus, with 10-20 collisions per event on average (across 45 μm TiO_2), these considerations taken together set the maximum energy budget of ~700 eV for detected events.

With this basic picture of the recoil electron transport in view, we can speculate as to the mechanism of the radiation detection, though the details are not yet entirely clear. First, it is important to note that the energy for oxygen vacancy production $E(V_O)$ is thought to be 3-5 eV in various forms of bulk TiO_2 [28]. So, individual scatterings of recoil electrons and energy deposited in the aggregate are sufficient to produce many vacancies in μ-thick devices. In their conclusions, the authors of [28] provide a possible interpretation of oxygen vacancy behavior in TiO_2-based RRAM devices, where the TiO_2 surface is in contact with an electrode which holds the Fermi energy in the material at ~0.4-1.4 eV below the bulk limit and in the O-poor limit, $E(V_O)$ can drop below 1 eV. This is consistent with our hypothesis for the radiation detection interaction, as the likelihood of Compton backscattering is greatest in the higher density electrode material. As [28] points out, this same region near the electrode is also where $E(V_O)$ drops low enough to be nearly spontaneous. The introduction of energy via Compton scattered recoil electrons may be powering vacancy production at the electrode-oxide interface and promoting the formation of conducting filaments under the sub-threshold bias applied in this work.

Adding further support for this interpretation for radiation detection in μ-thick TiO_2 memristors, our observations are also consistent with previously established radiation hardness of nano-thick devices of the same material [16-22]. In these works, electrodes are typically 20-30 nm thick, which greatly reduces the Compton scattering probability, relative to μ-thick devices, but more importantly, TiO_2 films in these devices are typically 20-30 nm bulk and have as little as a 5 nm O-poor layer at the interface. Given the mean free path of recoil electrons is in the range of several 100's nm, gamma

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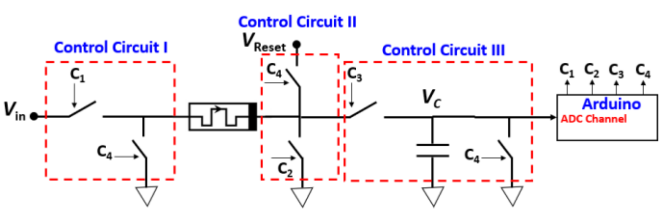
interactions in nano-thick devices will produce recoil electrons that transit the O-poor region and the entire film without any interaction. Thus, γ-ray interaction events in nano-thick devices do not efficiently produce vacancies or other displacement damages and consequently, do not produce detectable signals for a sensing application.

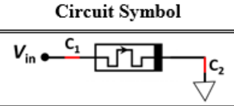
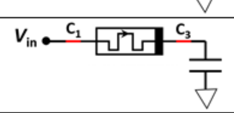
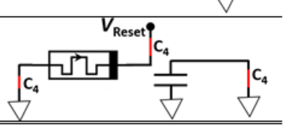
V. MemSens Circuit

A. Circuit Diagram

The results obtained in the preceding section are used to propose a standalone MemSens circuit. Fig. 7(a) shows the schematic diagram of the sensing circuit. The circuit consists of MemSens, six CMOS bilateral switches (CD4066B chip), a 1 µF capacitor and an Arduino board. The Arduino board generates the control signals for the switches ($C_{1\text{-}4}$). In addition, the board samples the capacitor voltage using its internal analog to digital converter (ADC). The switches C_1-C_4 are utilized to achieve three different operation modes. As demonstrated in Fig 7(b), during Monitoring mode, DC input voltage (i.e. 0.15 V) is applied for 345 s, which is the maximum switching time obtained in Fig. 6 during the irradiation test. Thus, if MemSens is exposed to radiation during this time it will be enough to switch ON the memristor, while it stays OFF in the absence of radiation. In Detection mode, the state of MemSens is reflected to the connected capacitor to be fully charged if turn-ON event occurred in Monitoring mode. Finally, MemSens is switched OFF through V_{Reset} and the capacitor is discharged during Reset mode. This design allows cyclability of the device during the absence of radiation exposure.

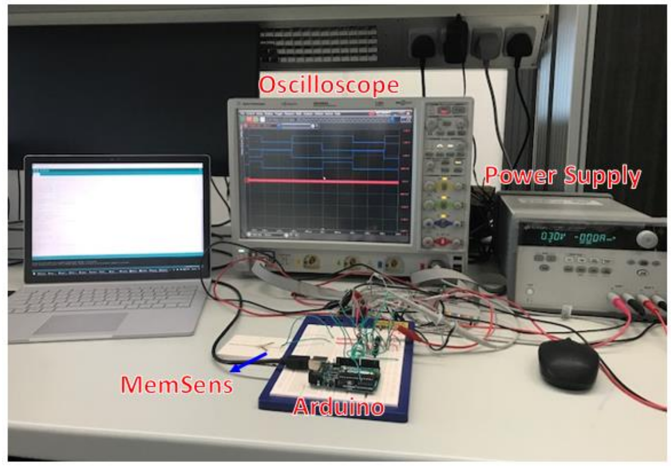


(a)

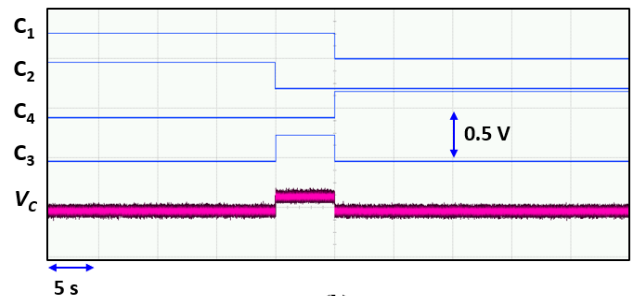
Mode	C_1	C_2	C_3	C_4	Circuit Symbol
Monitoring	1	1	0	0	
Detection	1	0	1	0	
Reset	0	0	0	1	

(b)

Fig. 7. (a) MemSens circuit schematic. (b) Testing modes for the proposed sensing circuit.



(a)



(b)

Fig. 8. (a) The built MemSens circuit. (b) Oscilloscope output during Detection and Reset modes.

The operation of the circuit is detailed as follows. Initially, all the control signals are turned OFF and the capacitor voltage (V_c) is zero. Now, C_1 and C_2 are turned ON to connect V_{in} to the memristor in a predetermined time window, i.e. 345 s. Then, C_2 is turned OFF and C_3 is turned ON to charge the capacitor. After 1 ms, the Arduino samples the capacitor's voltage, V_c, using its internal ADC. If the memristor was exposed to radiation, then V_c will be equal to V_{in}, otherwise, it is less than 10 mV. Finally, C_1 and C_3 are turned OFF and C_4 is turned ON. In this mode, the memristor is reset through passing a current in the opposite direction using V_{RESET} = 0.3 V. Further, the capacitor is discharged to ground for the next cycle evaluation. The memristor sensing approach described in Fig. 7 is built to achieve real time radiation detection prototype presented in Fig. 8 (a). In addition, Fig. 8 (b) shows the time domain measurement from the sensing circuit where the capacitor voltage is increased in Detection mode due to a radiation event.

B. MemSens Crossbar

In this section, the device-based sensing approach presented in Figs. 7 and 8 is used to propose a crossbar-based radiation detection methodology. As shown in Fig. 9 (a), the testing approach explained in the preceding section is applied

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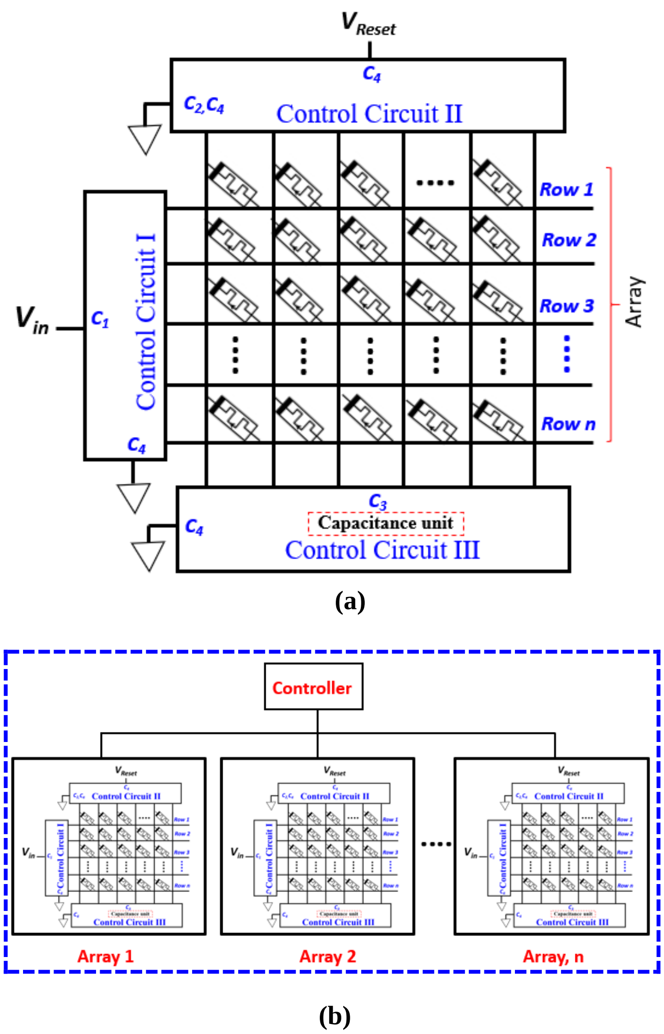

Fig. 9. (a) Proposed crossbar-based sensing circuit. (b) Multiplexed arrays-based sensing system which will allow multiple arrays to be in sensing mode at the same time.

consecutively to each memristive row. For instance, the input voltage V_{in} generated by the control circuit is applied to *Row* 1 to allow radiation detection to take place in this row. Having the detection carried by multiple devices (in the same row) at the same time increases the detection probability and consequently improves the system detection accuracy. As explained in section IV, the memristive devices fabricated in this work are radiation hardened under passive exposure. Therefore, the remaining memristive rows are not disturbed as long as their input voltages nodes are floating and hence, the subsequent row is activated once it is connected to the input voltage. Fig. 9 (b) suggests having multiplexed memristive arrays for improved utilization and increased accuracy of the proposed MemSens. For instance, while *Row* 1 in Array 1 is activated, the memristor devices may be exposed to radiation during Monitoring mode. However, the remaining time in this mode may not be enough to switch-ON the memristors and record the radiation event. Thus, it will be powerful to have other memristor rows in different arrays that are activated in subsequent (staggered) manner to allow relatively real-time detection. It is worth mentioning that the crossbar size and the number of the multiplexed arrays are decided based on the target application.

One of the key advantages of MemSens is the associated low power consumption during radiation detection. As explained earlier, a bias voltage of 0.15 V is required to operate MemSens. In the absence of radiation, the current passing through MemSens is in pico-range (10 pA) which results in a device power consumption of 1.5 pW. This increases to 30 µW when MemSens is exposed to radiation and consequently the current rises to 200 µA. For the proposed MemSens sensing circuit, an energy source of a common small coin battery can be used; which has a voltage of 2.9 V and capacity of 240 mAh [29]. Moreover, the circuit in Fig. 7 (a) can be designed with a power consumption of less than 10 µW; hence the maximum total power consumption for the total system is equal to 40 µW, which can be powered by energy harvesting system. To estimate the life time of the system, consuming 40 µW from a 2.9 V battery requires the battery to deliver 13.8 µA (40 µW/ 2.9 = 13.8 µA). With a battery capacity of 240 mAh, the life time of the system will be equal to 17391 hours (almost two years).

C. *MemSens and Existing Gamma Ray Sensors*

Gamma Ionizing radiation is extensively used in abundant applications such as environmental monitoring, medical imaging, radiation therapy, waste treatment, military, and customer products. Further applications include food irradiation which eradicates harmful organisms. However, radiation exposure to human body may have serious consequences, which can cause cell mutation and malfunction. Consequently, numerous γ-ray detectors are proposed and used for radiation protection and dosimetry assessment [30]. The development of environmental radiation monitoring and therapy have been influenced by the advancements of the semiconductor industry [31]. CMOS-based radiation detectors typically rely on the transit of radiation through a silicon-based active material, sensing the radiation events through a variety of mechanisms, and collection of the charge that is liberated, either directly (e.g., ionization) or indirectly (e.g. via electron band-gap promotion or charge trapping). Therefore, CMOS-based devices must be operated at relatively high voltage (up to 40V) for signal amplification, to perform charge integration which accounts for the majority of their power demands, in this case about 100 milliwatts.

There are two envisioned ways in which MemSens circuit can improve the state-of-the-art. First, MemSen circuit lowers power demands through the radiation interaction and registration mechanism in the device. In MemSens, the mechanism of radiation registration is primarily through ionic and molecular dissociation and defect formation, not charge production (direct ionization) and collection like in CMOS devices. The dissociations and defects in the memristor material improve the formation of current carrying filaments and change the resistive state of the device. Because a readout circuit needs only to register the resistivity change to capture the radiation event, it does not require significant

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amplification. Therefore, the anticipated reduction in power usage is in the range of ×100 - ×10,000 over that of the CMOS-based devices. Second, and consequently, the simplified circuit necessary for readout means that size and weight reductions around ×2 - ×3 are possible with significant effort on packaging development and design. Thus, this work presents a new class of high dense, ultra-low power radiation sensor such that many devices can be integrated into a single platform.

Regarding the sense in which MemSens is a real-time detection device, this technology does not represent superior capabilities to the best features of all existing approaches, but instead is capable of certain trade-offs. For instance, MemSens devices are all dose-integrating devices; they cannot resolve individual radiation interactions in time. So, the devices cannot be used to perform gamma spectroscopy and resolve different incident energies. For the same reason, the sense in which the detection is real-time is mainly for dosimetry applications, such as medical and personal protection [33], [34]. In this capacity, MemSens is much slower than chemical vapor deposition (CVD) diamond or Si-based real-time dosimeters, much faster than traditional thermoluminescent (TL) films (not faster intrinsically, but faster effectively, because of the electronic readout), but most attractively, potentially cheaper to produce and instrument than either of these technologies.

VI. Conclusion

This paper presented a novel microscale memristive stack, named MemSens, which consists of a $Ag/TiO_2/Cu$ device structure targeting sensing applications. MemSens exhibited bipolar switching behavior with improved power consumption, device area and endurance, compared to the existing microscale memristive devices. Although, nano-thick memristors were reported as radiation hardened, MemSens showed ability to sense 662 keV γ-rays through observable shifts in the obtained switching time of the device. Using this powerful property, a MemSens circuit was proposed in this work to allow for low power and relatively real-time radiation detection. Future work will include determination of minimum detectible dose, dose response, reproducibility and other features. This work is a milestone toward future advancement in RRAM-based memory, sensing and computing system.

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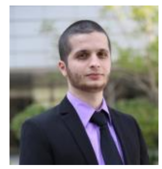

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