SAND2015-0316C

Overview of the Radiation Response of Anion-Based Memristive Devices

Michael L. McLain
Sandia National Labs
P.O. Box 5800, MS 1167,
Albuquerque, NM, 87185
505-844-6825
mlmclai@sandia.gov

Matt J. Marinella
Sandia National Labs
P.O. Box 5800, MS 1084,
Albuquerque, NM, 87185
505-844-7848
mmarine@sandia.gov

Abstract— **In this paper, we provide an overview of the current knowledge of radiation effects in anion-based memristive devices. We will specifically look at the impact of high dose rate ionizing radiation, total ionizing dose (TID), and heavy ions on the electrical characteristics of tantalum oxide (TaO_x), titanium dioxide (TiO_2), and hafnium oxide (HfO_x) memristors. The primary emphasis, however, will be placed on TaO_x memristors. While there are several other anion-based memristive devices being fabricated by the semiconductor community for possible use in valence change memories, most of the present radiation work has focused on one of these types of devices. There have also been numerous studies on radiation effects in cation-based chalcogenides such as germanium sulfides and selenides. However, that will not be discussed in this paper.**

TABLE OF CONTENTS

1. INTRODUCTION

The use of commercial off the shelf (COTS) non-volatile memory (NVM) technologies to store mission critical information has become commonplace in space and strategic systems. One of the primary drivers behind this evolution has been cost. While many of the commercially available memories surpass performance requirements in a normal operating environment, there is no guarantee that the memories will be able to meet requirements in a harsh radiation environment. Furthermore, the commercial industry is continually pursuing new technologies that will improve memory performance characteristics compared to the previous generation and/or technology. This is potentially problematic for designers that need radiation tolerant circuits because many of the emerging technologies have not been well characterized in radiation environments.

One possible candidate that has emerged as a possible flash memory replacement as Si-based technologies approach scaling limits is resistive random access memory (ReRAM) [1]. ReRAM (also known as redox or memristive memory or RRAM) is attractive to the electronic industry because of excellent scalability, high endurance, high switching speed, and low voltage operation [2-5]. Another benefit of ReRAM technologies is that most of the storage elements can be easily integrated into a back-end-of-the-line (BEOL) CMOS process which eliminates the need for an off-chip interface [6-10].

The storage elements within a ReRAM are two terminal metal-insulator-metal structures that are characterized by a low resistance on-state and a high resistance off-state depending on the electrical bias and bias history. These devices are often referred to as memristors or memory resistors. A schematic cross-section of the possible location of a tantalum oxide (TaO_x) memristor within the metallization layers of a silicon on insulator (SOI) technology is provided in Fig. 1. Being able to insert a memristor into the metallization layers is a key benefit for radiation applications because designers would be able to integrate the memory elements into a radiation-tolerant CMOS process. This eliminates possible radiation issues in the surrounding circuitry (e.g., access transistors). Indeed, many commercially available NVMs are fabricated using radiation-sensitive CMOS technologies [11], [12]. Furthermore, because of low write voltage requirements, ReRAMs do not require charge pump circuits. This is beneficial because charge pump circuits can be radiation sensitive [12].

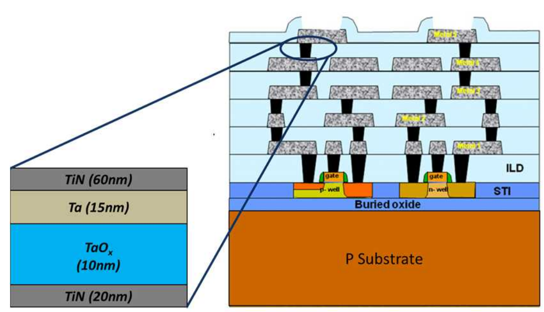

Figure 1. Schematic cross-section showing the possible location of a TaO_x memristor within the metallization layers of a SOI CMOS process (after [13], [14]).

978-1-4799-5380-6/15/$31.00 ©2015 IEEE

Memristive devices have been fabricated with a variety of different switching materials [6], [7], [10], [15], [16]. One possible way to categorize ionic switching devices is by the switching material (i.e., anion or cation devices) as described in [10]. Examples of the switching materials used in anion-based devices include the transition metal oxides (TMOs) tantalum oxide (TaO_x), titanium dioxide (TiO_2), and hafnium oxide (HfO_x) [6], [10]. Because the anion motion in these devices leads to valence changes of the metal, these devices are also called valence change memories [7]. Cation-based devices (also referred to as electrochemical metallization memory, conductive bridging RAM, or programmable metallization cells) typically have an electrochemically active electrode and use electrolytes such as germanium selenides and sulfides (i.e., chalcogenides) [7], [10], [15], [16]. For a complete description of the physics and operation of the different types of memristive devices refer to [6-8], [10], [15], [16]. The focus of this paper is on anion-based tantalum oxide (TaO_x), titanium dioxide (TiO_2), and hafnium oxide (HfO_x) memristive devices.

The potential use of ReRAM technologies in space or aerospace applications necessitates that we understand the susceptibility of memristive devices in various radiation environments. In this paper we will provide an overview of several studies that have investigated the impact of high dose rate ionizing radiation, total ionizing dose (TID), and heavy ions on the electrical characteristics of TaO_x, TiO_2, and HfO_x memristors [3], [4], [14], [17-30]. However, an emphasis will be placed on the response of TaO_x memristors. The paper is organized as follows: In the second section, a brief discussion on the operation of anion-based memristors is presented. After that, the results of several different ionizing radiation (dose rate and TID) and heavy ion experiments are provided. The fourth section discusses some of the observed radiation responses and the implication of the results. The paper is then concluded.

2. DEVICE OPERATION

As mentioned previously, memristive devices act as electrical resistance switches. In order to activate the switching property between a low and high resistance state, an initial electroforming cycle is generally required. Once a low resistance channel has been formed, the device can be reset into the high resistance off-state (HRS) and subsequently set to a low resistance on-state (LRS) by applying the appropriate field or forcing the appropriate current. Shown in Figure 2 are typical hysteretic I-V loops for a TaO_x memristor swept multiple times. For this hysteresis sweep, a positive current was forced to set the devices into a low resistance on-state and a negative voltage was applied to reset the devices into a high resistance off-state. A positive voltage can also be used to set the devices as described in [20], [29]. Generally, the bias (or current) is applied to the top electrode (anode) and the bottom electrode (cathode) is grounded. In the figure, the arrows indicate the direction of the sweep and the labels specify the state of the memristor (i.e., high resistance off-state or low resistance on-state). The moderate variation between consecutive hysteresis sweeps is typical of these research devices. The dashed box denotes the “read” window. Normally, a lower “read” voltage pulse is applied to the device to measure the resistance level. A read operation does not change the resistance state of the device. These I-V characteristics are similar to what is published in literature on other devices.

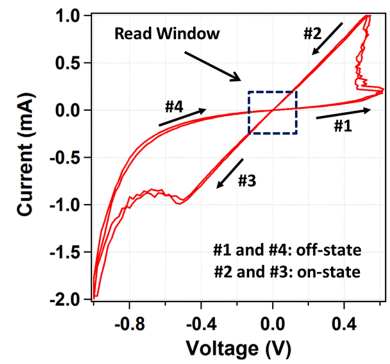

Figure 2. Representative electrical characteristics of a TaO_x memristor swept multiple times. The arrows indicate the direction of the sweep and the labels specify the state of the memristor [14].

The switching mechanism in anion-based devices involves redox (i.e., reduction and oxidation) reactions and the migration of oxygen anions (O^{2-}) or equivalently the positively charged oxygen vacancies [8], [10], [31]. The disassociation and transport of oxygen anions, which leads to chemical changes in the oxides, is thought to be due to electric and thermal fields [10]. For example, in TaO_x-based devices, these processes lead to the formation of a Ta-rich conducting filament of a certain radius when the device is set to a low resistance on-state [8], [31], [32]. Typically, the switching region is a localized conduction channel that is created in the electroforming process. A cross-sectional schematic of the formation (i.e., low resistance on-state) and dissociation of the filament (i.e., high resistance off-state) for a TaO_x device is shown in Figure 3. The voltage labels (i.e., V+ and V-) represent the externally applied bias on the anode, and the black arrows indicate the direction of the electric field. For more details on switching/transport mechanisms refer to [8], [10].

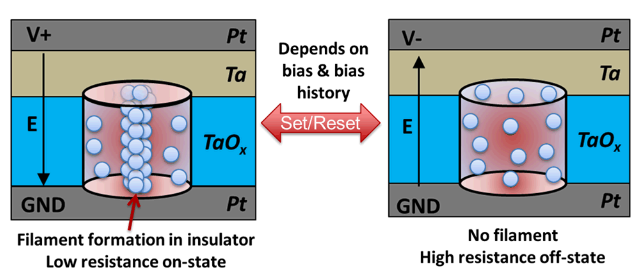

Figure 3. Illustration of the buildup and dissociation of oxygen vacancies (white circles) within the TaO_x layer. The voltage labels (i.e., V+ and V-) represent the externally applied bias on the anode (after [14], [31]).

3. RADIATION EFFECTS IN MEMRISTORS

The study of radiation effects in anion-based memristive devices has significantly increased in recent years. A subset of the different papers in literature that have investigated the radiation susceptibility of TaO_x, TiO_2, and HfO_x memristors is provided in Table 1. In the majority of the experiments, the devices were switched into the high resistance off-state prior to being irradiated. This state is generally considered to be the most radiation susceptible state. However, the devices were also occasionally tested in the low resistance on-state to ensure that there were not any unforeseen radiation vulnerabilities. In the following subsections we will review the radiation responses observed for the different memristive devices.

There were also several studies presented at the 2014 IEEE Nuclear and Space Radiation Effects Conference (NSREC) and recently published in the IEEE Transactions on Nuclear Science that looked at single event effects in memristors. This included several HfO_x papers as well as papers on commercial ReRAMs. However, these modeling and experimental efforts are beyond the scope of this paper.

Table 1. Radiation Studies on Memristors

Device Type	*Papers [references]*	*Radiation Environment*
TaO_x	[3], [4], [14], [17-19], [23], [33], [34]	Dose rate, TID, heavy ion
TiO_2	[4], [20-22], [24], [25]	TID, heavy ion
HfO_x	[26], [28-30]	TID, heavy ion

TaO_x-based Memristive Devices

As mentioned previously, the primary focus of this paper will be on TaO_x-based memristive devices. To date, the paper by McLain, et al. [14] is the only paper in literature that has looked at the effects of high dose rate ionizing radiation on memristive devices . In that paper, the focus was on two-terminal TaO_x memristors fabricated at Sandia National Laboratories (SNL). Transient data were obtained at a linear accelerator (LINAC) in electron beam mode for dose rates ranging from approximately $5.0x10^7$ rad(Si)/s to $4.7x10^8$ rad(Si)/s and for pulse widths ranging from 50 ns to 50 μs using. Shown in Figure 4(a) is a representative plot of the transient data obtained during a pulsed exposure for multiple devices under test (DUTs) on the same chip. The dose rate was 4.3×10^8 rad(Si)/s, the radiation pulse width was 500 ns, and the bias was -100 mV on the top electrode (anode). The bottom electrode (cathode) was connected to the 50 Ω terminator on the scope. As shown in the plot, DUT8 switched from a high resistance off-state (~11 kΩ) before irradiation to a low-resistance on-state (~160 Ω) after radiation. Radiation begins at time t=0 s. The other eight devices (not all shown in the figure) on this chip had a much smaller transient response and returned to their original pre-exposure levels. Indeed, the memristor response follows the radiation pulse shape and begins to recover after the pulsed exposure if there is not a state change. Note that the RF output of the LINAC was 5 μs wide which can be observed in the transient data. As would be expected, the device with the largest transient response (i.e., DUT8) was more susceptible to the high dose rate ionizing radiation. Despite the radiation-induced resistance change, this device could be reset into a high resistance off-state and was still fully functional following the radiation exposures. Further details on the experimental setup and results can be found in [14].

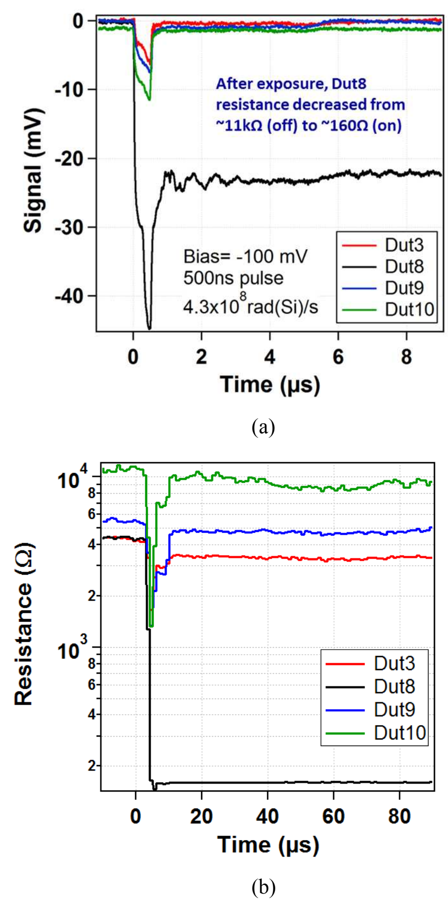

Figure 4. Transient response of (a) several DUTs at a dose rate of 4.3×10^8 rad(Si)/s and a pulse width of 500 ns. The anode of each device was biased at -100 mV during the exposure. The resistance versus time plot shown in (b) is for the same shot/devices in (a) [14].

Provided in Figure 4(b) is a plot of the resistance versus time for the same shot/devices shown in Figure 4(a). These resistances were calculated from data obtained on a scope with a higher voltage resolution. This was accomplished by splitting the signal to two scopes. Notice that the resistance level of both DUT3 and DUT9 changed after the radiation exposure; however, neither device changed states similar to DUT8. The enhanced susceptibility observed in some devices is still under investigation and is likely the key to determining the usefulness of these types of devices in a higher dose rate environment. Similar results were obtained for the other devices tested. More specifically, radiation-induced switching and resistance changes were observed in some of the devices at a nominal dose rate of 4.0×10^8 rad(Si)/s but not below 4.0×10^8 rad(Si)/s.

Static dose rate upset tests were also performed at a dose rate of ~$3.0\text{x}10^8$ rad(Si)/s using a pulsed X-ray machine [14]. The data suggest that the test condition influenced the radiation response. In particular, when the devices were floating and isolated during irradiation, some of the devices changed from a high resistance off-state to a low-resistance on-state. However, when the devices were grounded or shorted together but floating, a resistance change was not observed in any of the memristors. These results suggest that a power gating configuration is not the best option if using TaO_x memristors in dose rate environments. The power gating technique would allow control logic to turn off the power to a given memristor during an inactive period, effectively floating and isolating the terminals [35]. It is speculated that floating the terminals might be worst-case because of the lack of a discharge path. Similar to the LINAC results, there was variation in the dose rate response and all devices remained functional following the pulsed X-ray exposures.

In addition to using the LINAC for assessing the dose rate vulnerability of SNL-fabricated TaO_x memristors, the authors in [14] also used the LINAC to investigate TID effects. The primary purpose of those experiments was to determine if the dose received during the dose rate experiments was impacting the observed response. The data suggest that the cumulative dose does not impact the dose rate response. This can be seen in in Figure 5 for the pre- and post-exposure resistance versus accumulated dose for a single device tested at the LINAC. The resistances were calculated with transient data at nominal dose rate levels of 1×10^8 rad(Si)/s and 4×10^8 rad(Si)/s and pulse widths between 50 ns and 50 µs. As the data indicate, switching was observed at 4×10^8 rad(Si)/s but not at 1×10^8 rad(Si)/s. At the highest dose rate level, the response varied from device to device. It is interesting to note from the plot in Figure 5 that at higher dose levels, but lower dose rates, the DUT did not switch states or have a significant resistance change. However, at lower dose levels, but higher dose rates, the DUT switched states. This suggests that the dose rate (i.e., rate of energy deposition) is the cause of the resistance changes and not the dose. When the DUT was kept in the low resistance on-state during the radiation exposures, there was not a large change in the resistance as shown in the last two higher dose rate data points.

There have also been numerous other studies looking at the TID response of TaO_x memristors using 10 keV X-rays, Cobalt-60 γ-rays, and heavy ion sources as described in [3], [4], [14], [17-19], [33], [34]. In these studies the devices slightly differed (e.g., oxide layer thickness and/or contact materials). Overall, the results were very promising and

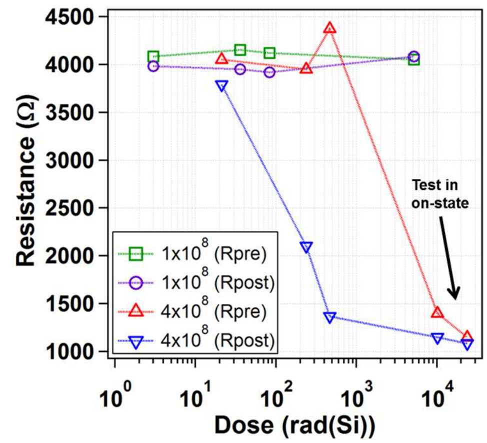

Figure 5. Plot of the pre- and post-exposure resistance versus dose for different dose rates. The data were obtained at the LINAC on a single device. In the last two exposures at the higher dose rate, the device was kept in the low resistance on-state [14].

suggest that there was minimal impact on the electrical characteristics until higher TID levels were reached. Similar to the LINAC experiments, devices set to the low resistance on-state before irradiation were tolerant to TID effects. However, when the devices were set to the high resistance off-state before irradiation, state changes were observed once a TID threshold was surpassed. In most of the previous work, the devices were floating during irradiation. Thus, electrical measurements were performed prior to and immediately following the TID exposures using a parameter analyzer.

There have also been studies characterizing devices that were biased or grounded during the radiation exposures (e.g., work by McLain, et al. [14], [33]). An example of one set of data is presented in Figure 6. Here, the resistances of a single TaO_x dog-bone device are plotted as a function of accumulated dose and applied bias. The step-stress exposures were completed consecutively using a Cobalt-60 γ-ray source. The resistance values were determined at a voltage of ~50 mV from in-situ read measurements performed immediately after each radiation exposure. As indicated by the data, the DUT had a radiation-induced resistance change when the step stress was 1 Mrad(Si). For lower step stress levels, the DUT did not change states. Furthermore, neither the bias nor the polarity of the bias appeared to impact the radiation response. The DUT could be reset into a high resistance off-state and was still fully functional following the exposures. There was, however, a slight shift in the off-state resistance after the first radiation-induced resistance change. This is still under investigation but is similar to the stress effects observed when trying to reset a device after an abnormally large bias is applied to set the device. What is also apparent in this work is that not every device is susceptible to TID even at higher levels.

Resistance data obtained for two dog-bone devices irradiated with a 10 keV X-ray Aracor source are shown in Figure 7 [33]. In this experiment, the devices were switched

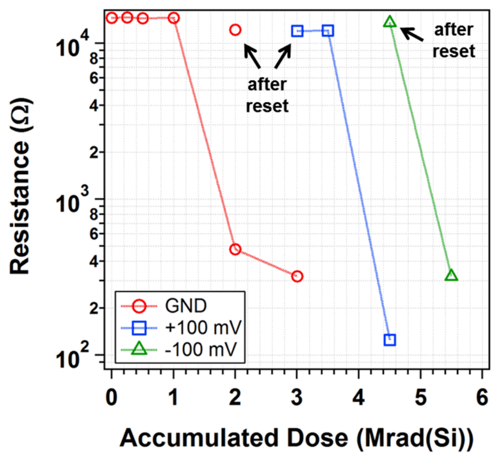

Figure 6. Plot of the resistance versus accumulated dose for three different biases (i.e., grounded and ±100 mV). The on-state resistance before irradiation was ~200 Ω. The device was reset after each observed radiation-induced state change [14].

into the high resistance off-state prior to irradiation. During the step stress exposures, all terminals were either grounded or floating. Note that before conducting the X-ray experiments, the devices were exposed to Si ions. In that experiment, the devices were switching resistance states at much lower fluence levels than expected. Because Si ion exposures will also cause high levels of ionization, it was postulated that the observed resistance changes in the devices were due to ionization. Therefore the devices were irradiated with the 10 keV Aracor source. It is certainly possible that the ion exposures impacted the TID response due structural changes in the memristive layer that are not apparent in the electrical characteristics, but the high TID threshold seemed to suggest that this was not the case. As observed in the data, there were minimal changes in the off-state resistance of DUT2 for three different runs. However, there were radiation-induced resistance changes in DUT1 depending on the step stress TID level and the bias condition during the experiment. It should be noted that DUT1 enters an intermediate state and not a low resistance on-state. One possible reason for this response is that the photoelectrons produced by the X-rays could be just a few keV (e.g., 1-5 keV). This would likely lead to a non-uniform deposition of charge (trapped or delayed charge) resulting in a non-uniform, transient electric field across the device that would impact the formation of a conducting filament.

The TID experiments conducted by Hughart, et al. and published in [4], [17], [34] also indicate that TaO_x dog-bone devices are not susceptible to TID until a TID threshold is surpassed. In some cases, tens of Mrad(Si) were required to switch the devices from a HRS to a LRS. In other cases, some of the devices switched after a few Mrad(Si). In either situation, these levels surpass those required for space or aerospace applications. Furthermore, Cobalt-60 gamma rays and 4.5 MeV protons did not change the resistances significantly at TID levels up to 2.5 Mrad(Si) and 5 Mrad(Si), respectively. Similar to what was observed in [33], floating the electrodes appears to make the device much more susceptible. This is apparent from the plot shown in Figure 8. More specifically, when floating the device, there is a resistance change around 100 krad(Si), and when shorting the electrodes together, the device did not change resistance states (up to ~11 Mrad(Si)).

The data obtained from the TID experiments indicate that it is possible for TaO_x devices to switch from a HRS to a LRS after a TID step stress threshold has been surpassed. If a read measurement is performed prior to reaching the charge threshold, the devices 'reset' back to a pre-irradiation state. This suggests that the devices do not have a cumulative TID effect. It is also apparent that these devices are minimally affected by step stress TID levels less than the TID threshold level at which the devices switch resistance states and that the threshold level varies from device to device.

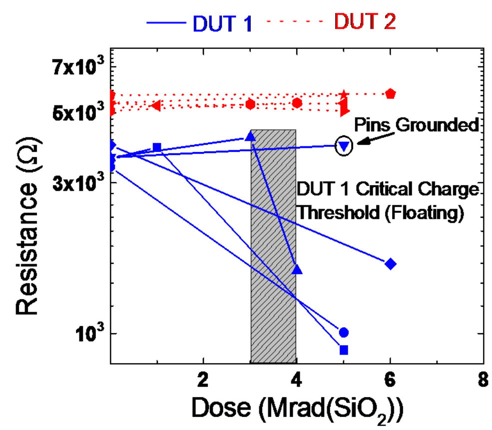

Figure 7. Plot of the resistance versus step stress TID level for devices tested with a 10 keV X-ray source. The resistance values were calculated at a voltage of ~50 mV. The in-situ read measurements were performed immediately after each shot [14].

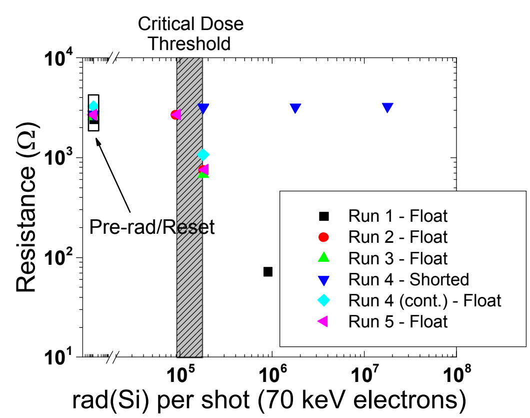

Figure 8. Resistance vs. rad(Si) per shot for a TaO_x memristor irradiated with 70 keV electrons. The part was reset between runs. The dose per shot threshold is ~100 krad(Si) in the floating case. When all pins are shorted, no change is observed for doses per shot up to 18 Mrad(Si) [34].

As described in [18], TaO_x memristors with an oxide thickness of 50 nm had large resistance changes after a dose of 180 krad(Si), but devices with a thickness of 25 nm had much smaller changes in resistance. This suggests that the TID response may depend on the thickness of the oxide film similar to what has been shown in silicon dioxide. Some of the work by Hughart et al., also seems to indicate that thicker devices are more susceptible to TID [4].

The heavy ion response of TaO_x devices is extremely promising. This is illustrated in Figure 9 with data from an experiment on a TaO_x memristor that was irradiated with 800 keV Ta ions. Here, switching was not observed until a vacancy concentration of $\sim 10^{19}$ cm^{-3} and an ion fluence of $\sim 10^{11}$ cm^{-2}. Also notice that the devices have a gradual reduction in resistance at high fluences. However, after irradiation, the devices remained fully functional. While some memristors show signs of cumulative damage after heavy ion irradiation, resetting the device multiple times may help restore degraded off-state resistances. Furthermore, continually resetting the device may gradually return the conduction channel region closer to its original state as discussed in [4], [34]. All of the heavy ion investigations suggest that the TaO_x technology is not susceptible to displacement damage effects for fluences less than 10^{10} cm^{-2}.

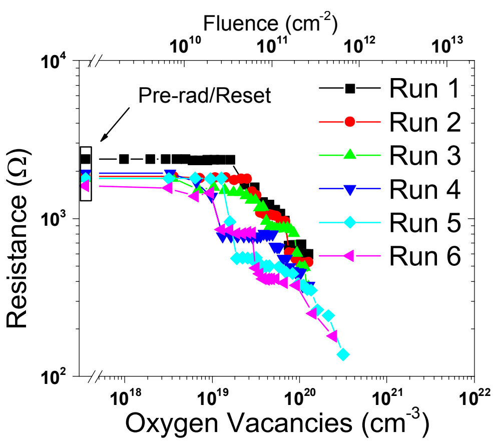

Figure 9. Resistance versus average oxygen vacancy concentration for a TaO_x memristor irradiated with 800 keV Ta ions. The part was reset between each run. The device was reset ten times after run four and twenty times after run five [4].

TiO_2 and HfO_x-based Memristive Devices

There are also numerous papers that have investigated the TID and heavy ion response of TiO_2 [4], [20-22], [24], [25] and HfO_x [26], [28-30] memristors. In general, the results are promising and suggest that there is minimal impact on the electrical characteristics until higher TID and fluence levels have been reached. An example of the TID performance of a TiO_2 technology is provided in Figure 10. Here we see that the TiO_2 memristor stays in the off-state after being irradiated whether the electrodes of the device are grounded or floating. The results provided in [21], [22] also indicate that TiO_2 memristive devices have a high tolerance to TID.

HfO_x-based memristive devices have also performed well in a TID environment. The work by Bi, et al., revealed that devices exposed to 10-keV X-ray irradiation did not have significant changes in resistance up to 7 Mrad(SiO_2) [28]. Furthermore, it was shown in [26], [29] that proton and γ-ray radiation did not have a major impact on the functionality, switching characteristics, or resistance until very high TID levels (i.e., greater than 5 Mrad).

With respect to heavy ions, there have been varying results presented in literature, but in general they have been positive. Shown in Figure 11 is a plot of the resistance versus the calculated concentration of oxygen vacancies created for a TiO_2 memristor irradiated with 800 keV Ta [4], [34]. The part was reset after each run. For the first three runs the resistance increases slightly (beginning around a fluence of 10^{11} cm^{-2}) and then decreases. The fourth run shows a significant drop in resistance at similar fluence levels. After run two, the device was cycled multiple times. During this process, the resistance degraded from 15 kΩ to 750 Ω after five cycles. Therefore, a larger reset current was applied, resulting in a higher off-state resistance ($R_{OFF} \approx 30$ kΩ at the start of Run 3). R_{OFF} did not show the same degradation after the remaining runs, but the variation increased. Refer to [4], [34] for more information on this experiment.

Both TaO_x and TiO_2 technologies show changes in resistance when exposed to 800 keV Ta ion irradiation. However, TiO_2 memristors show gradual increases in resistance with inconsistent decreases. The work by Barnaby, et al. and DeIonno, et al. also suggest that very high fluences of heavy ions are required to impact the electrical characteristics and the resistance of TiO_2-based memristive devices.

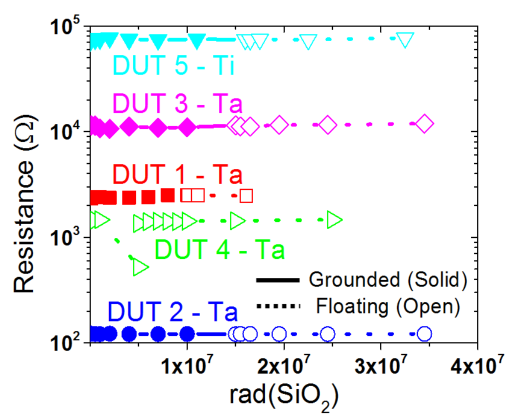

Figure 10. Resistance versus rad(SiO_2) for four TaO_x parts and one TiO_2 part. All parts (except DUT4) were set to the off-state. Solid lines indicate pins were grounded and dotted lines indicate pins were floating [4].

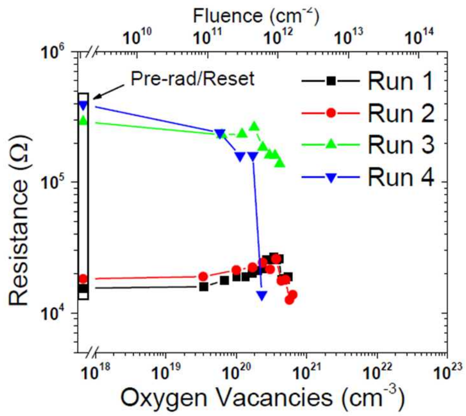

Figure 11. Resistance versus average oxygen vacancy concentration for a TiO_2 memristor irradiated with 800 keV Ta ions. The part was reset between each run. A higher reset current was used for runs three and four [4].

4. DISCUSSION

From the illustration in Figure 3, one can see how radiation-induced carriers generated by ionizing radiation could create local fields/currents in the memristive layer that would affect the operation of the device. More specifically, if radiation-induced carriers build up near the contacts, the internally generated field could impede the movement of the oxygen vacancies and the anions. If the polarization effect is large enough to create a field reversal, the device could switch resistance states. The transport of the radiation-induced carriers could also produce a transient current that is comparable to the current forced to set the devices into a low-resistance on-state. This could result in a state change as well. The reason that high dose rate ionizing radiation seems to be more of an issue in these devices compared to TID is probably due to the fact that more recombination occurs at longer times.

The increased susceptibility when floating the electrodes during a dose rate exposure is likely a consequence of the inability of generated charge to exit sensitive regions of the device (i.e., the TaO_x memristive layer). Intuitively, if the electrodes are floating and there is not a discharge path for the generated carriers, there may be an enhanced buildup of ionized carriers near the electrodes. This would create an internal field. If the internal field is comparable to the voltage required to set the device to a low resistance on-state, the device will change resistance states. Eventually the carriers will recombine, but because of the non-volatile properties of the memristive layer, the device will stay in the low resistance on-state once the device has switched states. When the terminals of the device are grounded, there is a discharge path for the generated carriers. Similar types of effects are also likely contributing to the TID response on a much smaller scale.

It is currently unknown what factors contributed to the increased vulnerability in certain devices. However, we speculate that the electroforming step stresses one device more than another. This could lead to a variation in the density of oxygen anions and vacancies, impact the formation, location, and number of filaments, and affect the overall integrity and reliability of the memristive layer. These differences would almost certainly be exacerbated by the addition of radiation-induced carriers. Another possibility is non-uniformity in the thickness of the memristive layer which is currently being investigated.

The anion-based devices discussed in this paper are likely insensitive to heavy ions due to the fact that they already have a large concentration of oxygen vacancies. For example, it is believed that TaO_x devices have an initial oxygen vacancy concentration on the order of 10^{18} to 10^{19} cm^{-3}. Therefore, the resistance of the devices will only degrade when a comparable amount of oxygen vacancies are introduced into the memristive layer in a localized channel region. This requires extremely high fluence levels that are not necessarily realistic in space or aerospace environments.

5. SUMMARY

In this paper, an overview of the effects of ionizing radiation and heavy ions on tantalum oxide (TaO_x), titanium dioxide (TiO_2), and hafnium oxide (HfO_x) memristive devices was presented. The radiation response of each type of device was promising in both TID and heavy ion environments. Despite observing state changes from the high resistance off-state to the low resistance on-state after a threshold (TID level or oxygen vacancy creation due to displacement damage in the memristive material), all devices were still functional following irradiation. Moreover, the level at which the dose or heavy ion damage impacted the devices was higher than what would be required for most space and aerospace applications. It was also observed that the experimental conditions impacted the radiation response of TaO_x devices. More specifically, when the terminals of the memristor were left floating during irradiation, it appeared that the devices were more susceptible and changed resistance states at much lower doses. This is likely a consequence of the inability of generated charge to exit sensitive regions of the device. It was also observed that different devices were more vulnerable than others to total ionizing dose and that the thickness of the memristive layer impacted the radiation response.

The effect of high dose rate ionizing radiation on TaO_x-based memristors was also investigated in this paper. The experiments were performed using a Scandiflash Flash X-ray source and a LINAC in electron beam mode. The pulsed dose rate experiments were the first dose rate studies performed on any type of memristive memory technology. The data indicate that at higher dose rates, it is possible for the devices to switch from a high resistance off-state to a low resistance on-state. Despite the radiation-induced resistance changes observed in some of the devices, all devices were still functional following the dose rate exposures. From the LINAC dose rate data, it was also determined that the dose rate (i.e., rate of energy deposition) was the cause of the resistance changes and not the dose. This is a significant finding. The data also revealed that the dose rate at which a device switches resistance states varies from device to device; the enhanced susceptibility observed in some devices is still under investigation. In addition to that, it was found that the experimental conditions impacted the radiation response. More specifically, when the terminals of the memristor were isolated and floating during irradiation, it appeared that the devices were more susceptible and changed resistance states at much lower dose rates. Overall, the radiation performance of anion-based memristors is promising and could potentially enable the discovery of a radiation-hardened nonvolatile memory technology to be used in aerospace and space applications. However, we must continue to investigate and better understand the deleterious effects of various types of radiation on these devices.

ACKNOWLEDGMENTS

This work was funded by Sandia's Laboratory Directed Research and Development program. Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

REFERENCES

[1] International Technology Roadmap for Semiconductors 2013 Edition, Emerging Research Devices. Available: www.itrs.net

[2] M.-J. Lee, C. B. Lee, D. Lee, S. R. Lee, M. Chang, J. H. Hur, Y.-B. Kim, C.-J. Kim, D. H. Seo, S. Seo, U. I. Chung, I.-K. Yoo, and K. Kim, "A fast, high-endurance and scalable non-volatile memory device made from asymmetric Ta_2O_{5-x}/TaO_{2-x} bilayer structures," *Nat. Mater.,* vol. 10, no. 8, pp. 625-630, July 2011.

[3] M. J. Marinella, S. M. Dalton, P. R. Mickel, P. E. D. Dodd, M. R. Shaneyfelt, E. Bielejec, G. Vizkelethy, and P. G. Kotula, "Initial assessment of the effects of radiation on the electrical characteristics of TaO_x memristive memories," *IEEE Trans. Nucl. Sci.,* vol. 59, no. 6, pp. 2987-2994, Dec. 2012.

[4] D. R. Hughart, A. J. Lohn, P. R. Mickel, S. M. Dalton, P. E. Dodd, M. R. Shaneyfelt, A. I. Silva, E. Bielejec, G. Vizkelethy, M. T. Marshall, M. L. McLain, and M. J. Marinella, "A comparison of the radiation response of TaO_x and TiO_2 memristors," *IEEE Trans. Nucl. Sci.,* vol. 60, no. 1, pp. 4512-4519, Dec. 2013.

[5] M. Marinella, "The future of memory," in *2013 IEEE Aerosp. Conf.*, Big Sky, MT, 2013, pp. 1-11.

[6] H. Akinaga and H. Shima, "Resistive random access memory (ReRAM) based on metal oxides," *Proc. IEEE,* vol. 98, no. 12, pp. 2237-2251, Oct. 2010.

[7] R. Waser, R. Dittmann, G. Staikov, and K. Szot, "Redox-based resistive switching memories – nanoionic mechanisms, prospects, and challenges," *Adv. Mater.,* vol. 21, no. 25-26, pp. 2632-2663, July 2009.

[8] F. Miao, J. P. Strachan, J. J. Yang, M.-X. Zhang, I. Goldfarb, P. E. Torrezan, R. D. Kelley, G. Medeiros-Ribeiro, and R. S. Williams, "Anatomy of a nanoscale conduction channel reveals the mechanism of a high-performance memristor," *Adv. Mater.,* vol. 23, no. 47, pp. 5633-5640, Nov. 2011.

[9] G. W. Burr, B. N. Kurdi, J. C. Scott, C. H. Lam, K. Gopalakrishnan, and R. S. Shenoy, "Overview of candidate device technologies for storage-class memory," *IBM J. Res. and Dev.,* vol. 52, no. 4/5, pp. 449-464, July/Sept. 2008.

[10] J. J. Yang, D. B. Strukov, and D. R. Stewart, "Memristive devices for computing," *Nat. Nano.,* vol. 8, no. 1, pp. 13-24, Jan. 2013.

[11] S. Gerardin and A. Paccagnella, "Present and future non-volatile memories for space," *IEEE Trans. Nucl. Sci.,* vol. 57, no. 6, pp. 3016-3039, Dec. 2010.

[12] S. Gerardin, M. Bagatin, A. Paccagnella, K. Grurmann, F. Gliem, T. R. Oldham, F. Irom, and D. N. Nguyen, "Radiation effects in flash memories," *IEEE Trans. Nucl. Sci.,* vol. 60, no. 3, pp. 1953-1969, June 2013.

[13] M. J. Marinella, A. J. Lohn, J. E. Stevens, S. Decker, P. R. Mickel, D. R. Hughart, B. Evans, and E. Debenedictis, "A CMOS compatible, forming free TaO_x ReRAM," presented at the ECS Fall Meeting, San Francisco, CA, 2013.

[14] M. L. McLain, H. P. Hjalmarson, T. J. Sheridan, P. R. Mickel, D. J. Hanson, J. K. McDonald, D. R. Hughart, and M. J. Marinella, "The Susceptibility of TaO_x-based Memristors to High Dose Rate Ionizing Radiation and Total Ionizing Dose " *IEEE Trans. Nucl. Sci.,* vol. 61, no. 6, pp. 2997-3004, Dec. 2014.

[15] M. N. Kozicki, C. Gopalan, M. Balakrishnan, P. Mira, and M. Mitkova, "Nonvolatile memory based on solid electrolytes," in *Non-Volatile Memory Tech. Symp.*, Orlando, FL, 2004, pp. 10-17.

[16] I. Valov, R. Waser, J. R. Jameson, and M. N. Kozicki, "Electrochemical metallization memories—fundamentals, applications, prospects," *Nanotechnology,* vol. 22, no. 28, p. 254003, May 2011.

[17] D. R. Hughart, S. M. Dalton, P. R. Mickel, P. E. Dodd, M. R. Shaneyfelt, E. Bielejec, G. Vizkelethy, and M. J. Marinella, "Total ionizing dose and displacement damage effects on TaO_x memristive memories," in *2013 IEEE Aerosp. Conf.*, Big Sky, MT, 2013, pp. 1-10.

[18] L. Zhang, R. Huang, D. Gao, P. Yue, P. Tang, F. Tan, Y. Cai, and Y. Wang, "Total ionizing dose (TID) effects on TaO_x-based resistance change memory," *IEEE Trans. Elec. Dev.,* vol. 58, no. 8, pp. 2800-2804, Aug. 2011.

[19] E. Atanassova, A. Paskaleva, R. Konakova, D. Spassov, and V. F. Mitin, "Influence of γ radiation on thin Ta_2O_5–Si structures," *Microelectronics Journal,* vol. 32, no. 7, pp. 553-562, July 2001.

[20] H. J. Barnaby, S. Malley, M. Land, S. Charnicki, A. Kathuria, B. Wilkens, E. DeIonno, and W. Tong, "Impact of alpha particles on the electrical characteristics of TiO_2 memristors," *IEEE Trans. Nucl. Sci.,* vol. 58, no. 6*,* pp. 2838-2844, Dec. 2011.
[21] E. DeIonno, M. D. Looper, J. V. Osborn, H. J. Barnaby, and W. M. Tong, "Radiation effects studies on thin film TiO_2 memristor devices," in *2013 IEEE Aerosp. Conf.*, Big Sky, MT, 2013, pp. 1-8.
[22] W. M. Tong, J. J. Yang, P. J. Kuekes, D. R. Stewart, R. S. Williams, E. DeIonno, E. E. King, S. C. Witczak, M. D. Looper, and J. V. Osborn, "Radiation hardness of TiO_2 memristive junctions," *IEEE Trans. Nucl. Sci.,* vol. 57, no. 3*,* pp. 1640-1643, June 2010.
[23] F. Tan, R. Huang, Z. An, Y. Cai, Y. Pan, W. Wu, H. Feng, X. Zhang, and Y. Y. Wang, "Investigation of the response of TaOx-based resistive random-access memories to heavy-ion irradiation," *IEEE Trans. Nucl. Sci.,* vol. 60, no. 6*,* pp. 4520-4525, Dec. 2013.
[24] E. DeIonno, M. D. Looper, J. V. Osborn, and J. W. Palko, "Displacement damage in TiO_2 memristor devices," *IEEE Trans. Nucl. Sci.,* vol. 60, no. 2*,* pp. 1379-1383, Apr. 2013.
[25] Y. Chi, R. Liu, Z. Tang, and R. Song, "Total ionizing dose effect on low on/off switching ratio TiO_2 memristive memories," *IEEE Trans. Nucl. Sci.,* vol. 61, no. 4*,* pp. 1889-1893, Aug. 2014.
[26] H. Xiaoli, W. Wei, B. Butcher, S. Tanachutiwat, and R. E. Geer, "Superior TID hardness in $TiN/HfO_2/TiN$ ReRAMs after proton radiation," *IEEE Trans. Nucl. Sci.,* vol. 59, no. 5*,* pp. 2550-2555, 2012.
[27] Y. Wang, L. Hangbing, W. Wang, Q. Liu, S. Long, W. Qin, H. Zongliang, Z. Sen, L. Yingtao, Z. Qingyun, L. Wentai, Y. Jianhong, and M. Liu, "Highly stable radiation-hardened resistive-switching memory," *IEEE Elec. Dev. Lett.,* vol. 31, no. 12*,* pp. 1470-1472, Dec. 2010.
[28] J. S. Bi, Z. S. Han, E. X. Zhang, M. W. McCurdy, R. A. Reed, R. D. Schrimpf, D. M. Fleetwood, M. L. Alles, R. A. Weller, D. Linten, M. Jurczak, and A. Fantini, "The impact of X-ray and proton irradiation on HfO_2/Hf-based bipolar resistive memories," *IEEE Trans. Nucl. Sci.,* vol. 60, no. 6*,* pp. 4540-4546, Dec. 2013.
[29] R. Fang, Y. Gonzalez Velo, W. Chen, K. E. Holbert, M. N. Kozicki, H. Barnaby, and S. Yu, "Total ionizing dose effect of γ-ray radiation on the switching characteristics and filament stability of HfO_x resistive random access memory," *Appl. Phys. Lett.,* vol. 104, no. 18*,* pp. 183507-183507-5, May 2014.
[30] W. G. Bennett, N. C. Hooten, R. D. Schrimpf, R. A. Reed, M. H. Mendenhall, M. L. Alles, B. Jinshun, Z. En Xia, D. Linten, M. Jurzak, and A. Fantini, "Single- and Multiple-Event Induced Upsets in HfO_2/Hf 1T1R RRAM," *IEEE Trans. Nucl. Sci.,* vol. 61, no. 4*,* pp. 1717-1725, Aug. 2014.
[31] P. R. Mickel, A. J. Lohn, B. J. Choi, J. J. Yang, M.-X. Zhang, M. J. Marinella, C. D. James, and R. S. Williams, "A physical model of switching dynamics in tantalum oxide memristive devices," *Appl. Phys. Lett.,* vol. 102, no. 22*,* pp. 223502-5, June 2013.
[32] J. P. Strachan, G. Medeiros-Ribeiro, J. J. Yang, M.-X. Zhang, F. Miao, I. Goldfarb, M. Holt, V. Rose, and R. S. Williams, "Spectromicroscopy of tantalum oxide memristors," *Appl. Phys. Lett.,* vol. 98, no. 24*,* pp. -, June 2011.
[33] M. McLain, D. Hughart, D. J. Hanson, and M. J. Marinella, "Effects of ionizing radiation on TaO_x-based memristive devices," in *2014 IEEE Aerosp. Conf.*, Big Sky, MT, 2014, pp. 1-9.
[34] D. R. Hughart, A. J. Lohn, P. R. Mickel, P. E. Dodd, M. R. Shaneyfelt, A. I. Silva, E. Bielejec, G. Vizkelethy, B. L. Doyle, M. T. Marshall, M. L. McLain, M. J. Marinella, and S. M. Dalton, "Radiation-induced resistance changes in TaO_x and TiO_2 memristors," in *2014 IEEE Aerosp. Conf.*, Big Sky, MT, 2014, pp. 1-11.
[35] H. Zhigang, A. Buyuktosunoglu, V. Srinivasan, V. Zyuban, H. Jacobson, and P. Bose, "Microarchitectural techniques for power gating of execution units," in *Proc. ISLPED*, Newport, CA, 2004, pp. 32-37.

BIOGRAPHY

Michael McLain *received a B.S. in Electrical Engineering from the University of Arizona in 2004 and a M.S. and Ph.D. in Electrical Engineering from Arizona State University in 2006 and 2009, respectively. He has been with Sandia National Laboratories in Albuquerque, NM since 2009 working in the Radiation Effects Sciences Department. At Sandia, he is leading the program studying radiation effects in Resistive Memory (ReRAM/memristors). He is also the subject matter expert on several projects studying the effects of radiation on materials, devices, circuits, and systems. He has been involved in the radiation effects field for over 10 years.*

Matthew Marinella *is a principal Member of the Technical Staff at Sandia National Laboratories: Matthew Marinella received the PhD degree in electrical engineering from Arizona State University 2008 under Prof. Dieter Schroder. From 2008 to 2010 he was a Technology Development Device Engineer with Microchip Technology in Tempe, AZ. Since 2010 he has been a Senior Member of the Technical Staff with Sandia National Laboratories Advanced Semiconductor Device Research Group. Dr. Marinella is currently leading the Resistive Memory (ReRAM/Memristor) Program at Sandia. He is also involved in several novel electron device research projects and co-leading the advanced power device research program.*