Inexpensive Fabrication of Sn-Ag-Cu/Cu:ZnO/Al/PET Memristive Devices through Unconventional Synthesis Techniques

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***Abstract*—In the last few decades there has been rapidly growing research interest in resistive switching devices which led to tremendous technological advancements. In this work, copper (Cu) doped zinc oxide (ZnO) thin film was deposited on Aluminum coated PET conductive sheets having a sheet resistance of 20 Ω. Soldering balls of 2 mm diameter consisting of 96.5% Sn, 3% Ag and 0.5% Cu alloy were dropped into this thin film to form the Sn-Ag-Cu/Cu:ZnO/Al/PET memristive devices. No additional annealing temperature was required as the heat transfer from 350 °C hot soldering ball onto the Cu:ZnO thin film was sufficient to anneal the thin film. The electronic characteristics of the device were obtained by employing a function generator and an oscilloscope instead of an expensive source meter. The elimination of redundant hot furnace (for annealing) and source meter (for I-V characterization) resulted in the improvement of total equipment expenses by ~96%.**

***Index Terms*—memristor, material science, resistive switching, non-volatile memory, nanoscale materials, oxide semiconductors**

I. Introduction

At present, there is a great need for electronic devices which operate at low power with high switching rates in order to process huge volumes of data [1, 2]. In contrast, conventional silicon-based dynamic Random-Access Memory (RAM), static RAM and flash memory have obnoxious issues such as inadequate switching speed, high power requirements and poor scalability which led to the renewed interest towards Resistive Switching (RS) devices [3]. Resistive switching devices are increasingly becoming vital entities in revolutionizing circuit design for the advancement of numerous electronic applications. RS is a physical phenomenon in which due to the exertion of the strong electric field or current there is a sudden change in the memristance of the active layer [4].

RS devices are Non-Von Neumann like architecture [5]. In typical computing systems, RS devices provide a high degree of parallelism [6–8]. Interestingly, besides non-volatile memory, RS device finds its uses in designing digital switches, flip-flops, logic gates, latches and memristive interconnections in field programmable gate array with low on-chip area, faster and low power consumption [9]. Moreover, RS have been utilized in analog electronic applications such as adaptive filters, tunable gain amplifiers and chaotic circuits [9]. Furthermore, recent developments in the field of artificial neural networks have shown the need for RS devices [10, 11].

The existence of missing fourth fundamental circuit element, which is a two-terminal passive device called memristor was first posited by Chua in 1971 [12]. In 2008, it was eventually realized in HP labs [13]. Since last decade, the memristor has been an intrigued alternate to the conventional Complementary Metal–Oxide–Semiconductor (CMOS) devices in electronic circuits [14]. By adapting the magnitude and polarity of external power supply, the memristor can further be tuned [15]. In various applications like neuromorphic computations, digital image processing, oscillator circuits, analog computers, signal recognition and digital logic systems, memristor finds its uses due to its inbuilt capability to store data in the form of memristance [14]. This memory capability of memristor has made revolutionary effects in the design of electronic circuits [14, 16].

From non-volatile memory to programming logic, the main attributes of memristor that makes it attractive for a number of applications are modest switching voltage (<3 V), high endurance (> 10^6 cycles), eminent on-chip area (~42%), faster switching rates (~5 ns), reduced power loss (~10^{-6} W), multi-bit (6 bits) operations and high information retention time (~10 years) when compared to the existing transistor technology [4, 17]. In this regard, recently, there have been an increase in the research interests to design novel RS based circuits and systems.

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The RS device conventional structure consists of an active layer squeezed in between two electrodes. The electronic resistance in RS device represents the data stored in it where a logic 0 indicates a low resistive state and logic 1 represents a high resistive state. RS property in oxide materials like Al_2O_3, ZrO_2, SiO_x, TiO_2 and Ta_2O_5 was first observed by Hickmott in 1962 [18]. The first $Au/SiO_2/Al$ based RS cell structure was reported by Simmons and Verderber in 1967 [19]. After three decades, the research on RS thin films was accelerated by the group of Ignatiev [20]. Subsequently, a 64-bit Resistive RAM array was developed using the RS-CMOS hybrid technology which contains a 500 nm CMOS technology and $Pr_{0.7}Ca_{0.3}MnO_3$ RS device [21].

Recent developments in the field of RS device have led to a renewed interest in adoption of RS memories by multi-national companies like HP Labs, Samsung and Infineon [1, 9]. From the important features of RS device stated above, several electronic materials were exploited to obtain RS properties. To date, there has been a considerable amount of research carried out which tends to focus on RS devices in which, ferroelectric [19], ferromagnetic [20], perovskite oxides [21, 22], organic semiconductors [23], Transition-Metal Oxides (TMOs) [3] and polymers [4] were used as active material. By considering several optimistic features like easy fabrication, low operating voltage, power reduction, improved scalability, compact integration and coexistence with widely used CMOS devices [24] and faster switching rates, ferroelectric materials and TMOs were most preferred.

A considerable amount of literature has been shown that the outstanding electrical and structural attributes were provided by TMOs including ZnO [25], HfO_2 [26], and TiO_2 [27–29]. After rigorous experimentation, researchers preferred ZnO as an active material [30, 31] due to its simple synthesis procedure, reduced processing temperature, built-in availability of oxygen deficiencies and ions. In order to amend the electrical conductivity, defects, oxygen vacancies, and ions, several attempts were made to promote the functionalities of the device by inserting the metal impurities into ZnO thin film [32]. To inscribe this, different metal dopants such as copper (Cu) [32], cobalt [33], titanium [29], vanadium [30], lithium [31], aluminium [32] and lanthanum [33] were studied. Among these impurities, copper (Cu) contains an electron shell which is analogous to Zinc (Zn) spawning it to conveniently accommodate into the ZnO thin film structure [25]. Central to the discipline of RS devices, an extremely important active layer for memristive applications is Cu:ZnO because in ZnO, Cu acts as an electron trap, increases resistivity, and makes ZnO to act as a ferroelectric material. Thus, in summary, it is shown that Cu:ZnO is extremely preferred as active layer for RS devices with suitable electrodes [3, 34–40].

II. Materials and Methods

The economical synthetic methods were employed for blending the Cu:ZnO thin film. To begin with, 657.2 mg of zinc acetate di-hydrate was supplemented with 0.3 mL of di-ethanolamine and 10 mL of isopropanol. The resultant solution was amalgamated for 10 min at room temperature. Subsequently, 29 mg copper (II) acetate monohydrate was added to the resultant zinc acetate ensued by blending for 120 min at 330 K. This final solution was aged for 24 h. In our previous work [4], the physical characteristics of Cu:ZnO thin film were presented in detail.

III. Results and Discussions

Over the past few decades, the Al/PET conductive sheets are manufactured in bulk, widely used in food packaging applications and are available for negligible costs [39]. In this work, the solution of Cu:ZnO was dip-coated onto the Al/PET conductive sheet having a sheet resistance of 20 Ω. Thereafter, the deposited thin film was dried for 24 h at room temperature. As a result, ~50 nm Cu:ZnO thin film was deposited on Al/PET conductive sheet. It is important to mention that 96.5% Sn, 3% Ag and 0.5% Cu alloy is widely being used as soldering material. In order to reduce fabrication expenses through utilizing this already available material as electrode, soldering balls of 2 mm diameter consisting of this alloy were dropped onto the Cu:ZnO thin film to form the Sn-Ag-Cu/Cu:ZnO/Al/PET memristive devices shown in Fig. 1, where soldering ball was employed as top electrode and Al/PET conductive sheet acted as a bottom electrode with Cu:ZnO thin film as the active layer.

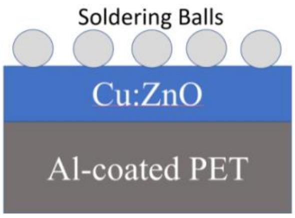

Fig. 1. Schematic of the fabricate d Sn-Ag-Cu/Cu:ZnO/Al/PET device architecture. Copper (Cu) doped zinc oxide (ZnO) thin film was deposited on aluminium coated PET conductive sheets having a sheet resistance of 20 Ω. Soldering balls of 2 mm diameter consisting of 96.5% Sn, 3% Ag and 0.5% Cu alloy were dropped into this thin film to form the Sn-Ag-Cu/Cu:ZnO/Al/PET memristive devices.

It is important to mention that the temperature of soldering balls was ~350 ℃. When these balls were dropped into the deposited thin film, heat transfer took place from the soldering balls to the Cu:ZnO thin film. This transfer was sufficient to anneal the Cu:ZnO thin film; no additional hot furnace was essential to anneal the thin film. The electronic characteristics of the device were obtained by employing a SM5060-2 function generator and a Scientific 30 MHz SM410 oscilloscope instead of an expensive source meter. The fabricated Sn-Ag-Cu/Cu:ZnO/Al/PET RS device is connected to the current to voltage converter which is depicted in Fig. 2. The function generator was employed to generate a sine signal of 2 V amplitude and 25 Hz frequency. On the other side, the Cathode Ray Oscilloscope (CRO) was employed to obtain the electronic device characteristic plots. In order to obtain these features, the current through the RS device was modified into a voltage signal through the employment of a current to voltage converter.

It is noteworthy to mention that ~10 V was supplied to each to the inverting and non-inverting terminals of the operational amplifier of the converter circuit. This converted signal was provided at the first channel (Y), nevertheless, the voltage across the memristive device was supplied to the second channel (X) of CRO which was engaged in its X-Y mode to obtain the room temperature *I-V* characteristics of the proposed RS device. The pinched hysteresis loop which indicates the signature of the memristor was obtained on the CRO as showcased in Fig. 2. Similar results were obtained through simulation roots.

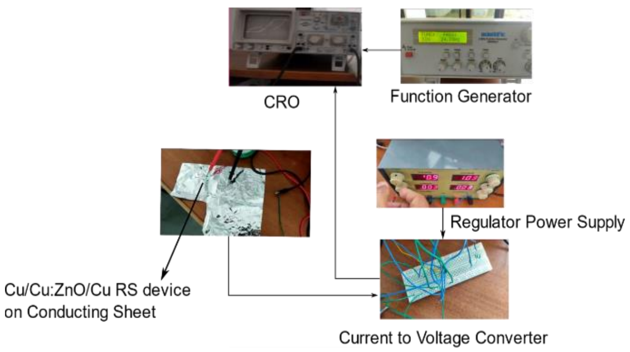

Fig. 2. Fabricated Sn-Ag-Cu/Cu:ZnO/Al/PET RS device connected to the current to voltage converter circuit. Function generator was employed to generate the sinusoidal signal whereas regulated power supply was engaged to supply the biasing voltage which is essential for the operational amplifier. The oscilloscope was functioned in its X-Y mode of operation.

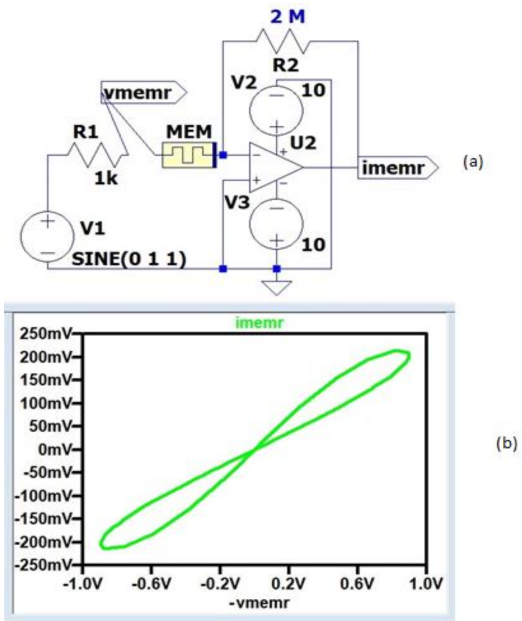

Fig. 3. (a) Circuit to convert current through memristor into the corresponding voltage signal, (b) and its plot on an oscilloscope. A 1 V and 1 Hz sinusoidal signal was applied as an input to memristor MEM through resistor R1. The operational amplifier U2, resistance R2, voltage sources V2 and V3 were utilized to convert the current through the memristor into the appropriate voltage signal. The plot between the memristive voltage -v_{memr} and the output voltage at i_{memr} depicted a pinched hysteresis graph.

Fig. 3(a) presentations the spice simulation of circuit which was employed to convert current through memristor into the corresponding voltage signal. A 1 V and 1 Hz sinusoidal signal was supplied to this circuit. The resistor R1 was engaged to defend the memristor MEM while the operational amplifier U2, resistor R2, voltage sources V2 and V3 were hired to modify the memristive current into the proportional voltage signal. The memristive voltage was represented as v_{memr} while the memristive current (in the form of voltage) was presented as i_{memr}. The mathematical relationships of the *I-V* curve were given by the following equations:

$$i_{memr} = 0.003 - 0.15v_{memr} + 0.007v_{memr}^2 \quad (1)$$

$$i_{memr} = -0.004 - 0.43v_{memr} + 0.21v_{memr}^2 \quad (2)$$

$$i_{memr} = 0.003 - 0.41v_{memr} + 0.18v_{memr}^2 \quad (3)$$

$$i_{memr} = -0.005 - 0.12v_{memr} + 0.11v_{memr}^2 \quad (4)$$

It is noteworthy to mention that Eq. (1) was obtained for the rising part of the curve along the positive i_{memr} axis, whereas Eq. (2) corresponded to the falling part of the curve in the first quadrant. It is important to mention that i_{memr} is the output terminal and thus the output voltage is at i_{memr}. Kindly note that i_{memr} represents the memristive current which was converted into the voltage at output through current to voltage converter. Consequently, Eq. (3) and Eq. (4) represent the rising and falling parts of the i_{memr} curve in the third quadrant.

The general form of the above equations is given as

$$\mathbf{i}_{memr} = \mathbf{X} + \mathbf{Y}v_{memr} + \mathbf{Z}v_{memr}^2 \quad (5)$$

The abovementioned relations can be vectorized into a lookup table for the modelling and simulation purpose as shown in Table I.

TABLE I: 2-D VECTORIZED REPRESENTATION OF THE I-V CHARACTERISTICS

Quadrant	X	Y	Z
Q1 rise	0.003	−0.15	0.07
Q1 fall	−0.004	−0.43	−0.21
Q3 rise	0.003	−0.41	0.18
Q3 fall	−0.005	−0.12	−0.11

As disclosed in Fig. 3(b), the $-v_{memr}$ versus i_{memr} plot was found to be a pinched hysteresis loop which represents the signature of the memristor, analogous to the *I-V* curve of the fabricated device depicted in the CRO of Fig. 2. This confirms that the Sn-Ag-Cu/Cu:ZnO/Al/PET memristor was successfully fabricated and characterized using inexpensive fabrication techniques through the elimination of redundant hot furnace (for annealing) and source meter (for *I-V* characterization). It is well known in literature that memristor exhibits randomness in its device characteristics [41]. However, electronic circuit design was efficiently performed such that the effects of randomness were mitigated in the overall performance of the memristive circuit [42, 43]. Furthermore, the memristance varies with temperature dependencies and other circuit elements connected to it, in addition to its stochastic behaviour [42, 43]. However, these drawbacks which are evident at the device level have been neutralised at the circuit level design [42, 43]. The proposed techniques are also valid even for other models of CRO & function generator as long as the signals generated and measured are within the specified range of these equipment. It is important to mention that these model variations do not adversely affect the memristive characteristics [43].

Based on the experimental data shown in Fig. 4, the memristor exhibited switching time dependent endurance property of $>10^5$ cycles. Grounded on the prolonged switching characteristics, it was identified that the memristor exhibited data retention time of >10 years. Therefore, the proposed memristor exhibited favorable characteristics for data storage applications.

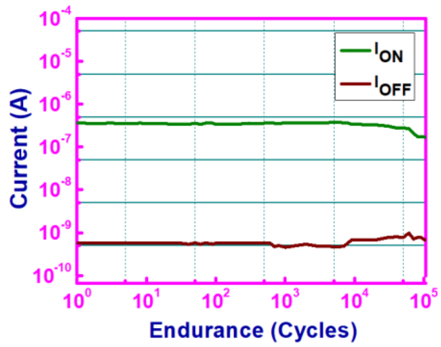

Fig. 4. Endurance of the proposed memristor. The Endurance was found to be stable for $>10^5$ cycles.

When compared to the traditional synthesis methods [1, 4, 17, 22, 40], the techniques presented in this work resulted in ~96% improvement of total equipment expenses. Such improvements were primarily owing to the employment of unconventional device characterization techniques. Furthermore, it is interesting to note that the device was fabricated and validated without the employment of clean room facilities.

IV. Conclusion

In summary, the proposed Cu:ZnO thin film was fabricated upon Al/PET conductive sheet. Soldering balls were employed as top electrode and the conductive sheet as the bottom electrode with Cu:ZnO thin film as the active layer in the proposed Sn-Ag-Cu/Cu:ZnO/Al/PET RS device. The utilization of the oscilloscope and function generator in order to obtain the *I-V* characteristics and the employment of heat from soldering rod instead of expensive hot furnace resulted in the improvement of total equipment expenses by ~96%. Importantly, the device was successfully fabricated and validated without the employment of clean room facilities. The ideas presented in this work pave the way for the futuristic inexpensive synthesis of memristive integrated circuits and systems.

Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

Panati S. Sreenivas Reddy, Michael Preetam Raj and Sravan K. Vittapu conducted the research; Panati S. Sreenivas Reddy, P. Michael Preetam Raj and Sravan K. Vittapu analyzed the results during experimentation; Panati S. Sreenivas Reddy wrote the paper; K. Senthil Kumar has given valuable inputs in every stage of the research; all authors had approved the final version.

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