

**INVESTIGATION OF SILVER OXIDE  
NANOPARTICLES IN POLYMANNOSE THIN  
FILM ON RESISTIVE SWITCHING  
CHARACTERISTICS**

**AU YONG HUEY LEEN**

**UNIVERSITI SAINS MALAYSIA**

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UNIVERSITI SAINS MALAYSIA**

**INVESTIGATION OF SILVER OXIDE NANOPARTICLES IN  
POLYMANNANOSE THIN FILM ON RESISTIVE SWITCHING  
CHARACTERISTICS**

By

**AU YONG HUEY LEEN**

**Supervisor: Prof. Ir. Dr. Cheong Kuan Yew**

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## DECLARATION

I hereby declare that I have conducted, completed the research work and written the dissertation entitled “**Investigation of Silver Oxide Nanoparticles in Polymannose Thin Film on Resistive Switching Characteristics**”. I also declare that it has not been previously submitted for the award for any degree or diploma or other similar title of this for any other examining body or University.

Name of Student: AU YONGHUEY LEEN

Signature:

Date: 8 AUGUST 2022



Witness by

Supervisor: PROF. IR. DR. CHEONG

Signature:

KUAN YEW



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## LIST OF ABBREVIATIONS

AFM	Atomic Force Microscopy
CNP	Cellulose nanofiber paper
CPU	Central Processing Unit
DRAMs	Dynamic random-access memories
ECM	Electrochemical metallization mechanism
EDX	Energy Dispersive X-ray
FTIR	Fourier Transform Infrared Spectroscopy
HRS	High resistive state
HRTEM	High Resolution Transmission Electron Microscopy
IGZO	Indium gallium zinc oxide
ITO	Indium tin oxide
LGB	Lophatherum gracile Brongn
LRS	Low resistive state
MIM	Metal/Insulator/Metal
PVA	Polyvinyl alcohol
RAMs	Random-access memories
ReRAMs	Resistive random-access memories
SCLC	Space-charge-limited-conduction
SEM	Scanning Electron Microscopy
SPA	Semiconductor Parameter Analyzer
SRAMs	Static Random Access Memory
TMV	Tobacco Mosaic Virus
WORM	Write Once Read Many

QDs

Quantum dots

IoTs

Internet of Things

## LIST OF APPENDICES

Appendix A     Sigma Aldrich IR spectrum table

Appendix B     Energy table for EDX analysis

## ABSTRAK

Filem tipis polimanosa telah menunjukkan sifat-sifat luar biasa yang penting dalam aplikasi ingatan capaian rawak (ReRAM). Beberapa sifat ini termasuk nisbah ON/OFF yang tinggi, tegangan READ yang relatif rendah, siklus daya tahan tinggi dan waktu retensi yang lama. Untuk alasan ini, filem tipis polimanosa dikatakan sebagai bahan bio-organik yang berpotensi dalam mnghasilkan sinapsis buatan untuk generasi berikutnya. Namun, prestasi pensuisan rintangan dalam kerintangan filem tipis polimanosa yang dimuat dengan nanopartikel logam belum dieksplorasi. Oleh itu, tujuan penelitian ini adalah untuk menguji kesan konsentrasi nanopartikel perak oksida yang ditambah dalam filem tipis polimanosa. Peranti dapat difabrikasi dengan mencampurkan D-mannose dan etanol sebagai prekursor dengan konsentrasi nanopartikel perak oksida yang berbeze iaitu 0 wt%, 0.5 wt%, 1.0 wt%, 2.0 wt% dan 10 wt%. Larutan prekursor akan disulutkan pada slaid kaca ITO dan dibiarkan kering selama 7 jam pada suhu 160°C untuk membentuk filem tipis kerintangan. Setelah peranti difabrikasi, memori bacaan dan rasio arus ON/OFF setiap peranti akan diuji secara terperinci. Kajian menunjukkan peranti dengan polimanosa yang dicampurkan dengan 1 wt% AgO NPs menunjukan prestasi terbaik dengan memori bacaan 0.65 V dan nisbah arus ON/OFF yang tinggi sebesar  $3.65 \times 10^2$  pada tegangan baca rendah (0.5 V). Pengetahuan yang diperoleh dari projek ini sangat bermanfaat untuk pengembangan ReRAM pada masa depan.



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**ABSTRACT**

Polymannose thin film has shown remarkable properties that are important in resistive switching random-access memory (ReRAM) and multistate switching memory applications. Some of these properties include a high ON/OFF ratio, relatively low READ voltage, high endurance cycle and long retention time. For these reasons, polymannose thin film is said to be a potential bio-organic material in producing the next-generation artificial synapses. However, the resistive switching behaviour of polymannose thin film loaded with metallic nanoparticles has not yet been explored. Therefore, this study aims to examine the effect of different concentrations of silver oxide nanoparticles incorporated in the polymannose thin film. The device can be fabricated with D-mannose powder and ethanol as precursors with different concentrations of silver oxide nanoparticles added which are 0 wt%, 0.5 wt%, 1.0 wt%, 2.0 wt% and 10 wt%. The precursor solutions were drop cast on the film and left dried for 7 hours at 160°C to form a resistive switching thin film. After the device is successfully fabricated, the read memory window and ON/OFF current ratio of each device were studied in detail. It is found that devices with polymannose loaded with 1 wt% AgO NPs demonstrated the best performance with a read memory window of 0.65 V and a high ON/OFF current ratio of  $3.65 \times 10^2$  at low read voltage (0.5 V). The knowledge gained from this work will be highly beneficial for the future development of the ReRAM.

# CHAPTER 1

## INTRODUCTION

### 1.1 Background of the study

According to Moore's law, the miniaturization of data storage is progressing at an exponential rate. A high data storage density and low energy consumption memory device are required to keep up with such rapid development. When searching for the ideal memory device, memristors have made the cut due to their non-volatility and wide range of applications. In recent years, researchers have put efforts into developing a bio-organic based ReRAM to further improve the sustainability and biodegradability of the device. Various approaches have been made including hybridization of two bio-organic materials, incorporation of nanomaterials, etc. to achieve the desired resistive switching properties. In this chapter, an introduction regarding the neuromorphic computing application, bio-organic based memristors, resistive switching mechanism and nanomaterials will be presented.

#### 1.1.1 Neuromorphic computing application

In this modern era, our world is being digitalized with advancements in technology at a rapid pace. This gave us an insight that electricity is going to be the main resource in the future. With that said, we must create a sustainable method to reduce electrical waste. The goal is to create an artificial synapse for neuromorphic computing applications. Artificial synapse emulates the biological synaptic signals in neuromorphic systems to achieve computation and autonomous learning behaviors like the brain. In the von-Neumann architecture, instruction fetches, and a data operation cannot occur at the same time in any stored-program computer. The central processing unit (CPU) is separated from the memory unit as shown in Figure 1.1. This is referred as the traffic

bottleneck and limits the performance of the system (Markgraf, 2007). Thus, artificial synapses are being studied to replace the traditional CPUs and Static Random-Access Memory (SRAMs) to artificial electronic neurons and synapses.

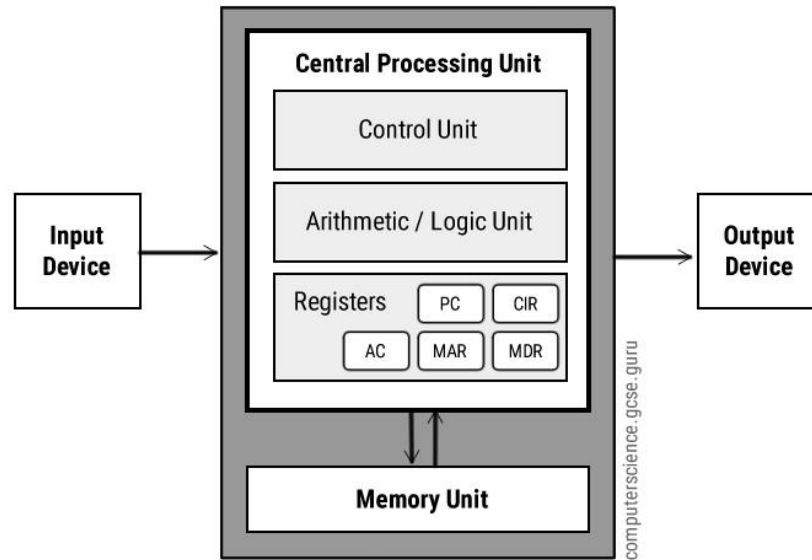


Figure 1.1 von-Neumann Architecture (Anon, 2019)

### 1.1.2 Bio-organic materials as insulator in memristor

A memristor is created acting as a synapse unit and the fundamental non-linear circuit element uses in computing and computer memory often referred as the Resistive Random-Access Memory (ReRAM). The ReRAM is a resistive switching memory proposed as a non-volatile memory where the information can be retained even if the power is removed. One of the main properties of non-volatile memory is its resistive switching behavior. Resistive switching is the ability to alter high resistance to low resistance. An ideal device would have a high ON/OFF ratio ( $>10$ ) with a low read voltage ( $V_{\text{read}}$ ) ( $<0.6$  V). The ON/OFF ratio is the ratio of the on-state and off state current without any applied gate voltage ( $V_g$ ). When a device has a high ON/OFF ratio it means that the device shows low current leakage indicating that it has a good digital performance.  $V_{\text{set}}$  is the voltage when the transition from OFF-state to ON-state occur

which serve as the “writing” process of the memory devices;  $V_{\text{reset}}$  is the voltage when current drop due to the transition from the ON-state back to the OFF-state which is also the “erasing” process of memory devices (Sivkov et al., 2020).

From previous research, bio-organic materials such as chitosan, silk or ferritin have gained interest to be used as a sustainable material for the future ReRAM. Recently, aloe polymannose has gained attention from researchers in search of a biodegradable and biocompatible green material. Aloe polymannose is a polysaccharide extracted from acemannan present in the inner leaf gel of the Aloe Vera plant. An elementary memristor can be simply represented as a metal-insulator-metal (MIM) structure where an electrochemical active material will be applied as the top electrode such as silver (Ag), aluminium (Al) or copper (Cu) and usually an inert metal such as gold (Au) or palladium (Pd) will be used as the bottom electrode. The polymannose come into the picture when it is used as the insulator layer in the MIM structure as shown in Figure 1.2.

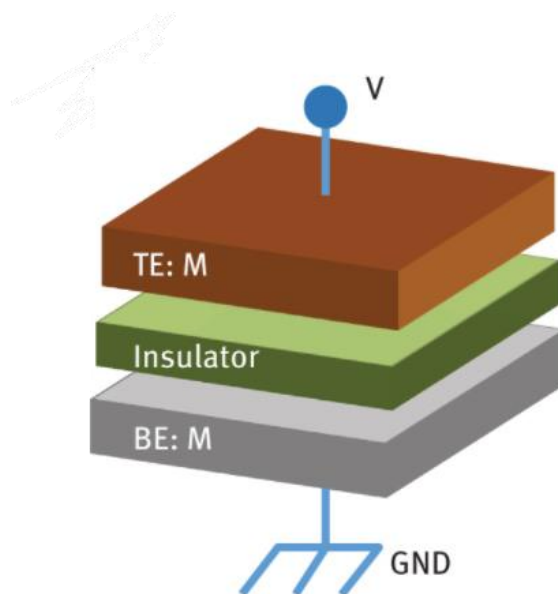


Figure 1.2 Typical metal (top electrode)-insulator-metal (bottom electrode) (MIM) structure of ReRAM with electrical biasing, (Prakash et al.,2016)

It is important to remember that the transportation of mobile charges across the MIM structure is dependent on the applied voltage (Lim et al., 2016), the inclusion of

metallic nanoparticles/ions (Lim et al., 2018) and the type of electrodes used (Hernández-Rodríguez et al., 2013). From previous studies, it is reported that polymannose displays good resistive switching behavior and is one of the potential bio-organic materials to build a sustainable ReRAM in near future (Tayeb et al., 2021). To fully understand and uncover the many possibilities of polymannose, the addition of metallic nanoparticles in polymannose can be one of the parameters to examine its effect on the resistive switching behavior of the device.

### **1.1.3 Resistive switching mechanism**

Different resistive switching mechanisms have been reported based on various parameters applied to the device. Based on a two-terminal junction with a metal/dielectric/metal structure, the resistive switching mechanism applied is usually driven by: electrochemical reaction, phase changes, tunnel magnetoresistance or ferroelectricity. Electrochemical redox reactions are mostly found in non-crystalline dielectrics sandwiched between electrodes. In this case, it is most likely to be applied to this study as non-crystalline bio-organic materials are being used as an insulator in the MIM device. Ion migration and redox reaction are the main processes involved in this mechanism and are affected by the electric potential, chemical potential, and temperature gradients over the reaction coordinates (Wang et al., 2020).

These mechanisms can be identified via the replotting of the J-V characteristics in the logarithmic scale. The experimental data can be fitted into different laws. Ohm's law indicates the domination of Ohmic conduction while the combination of Ohm's and Child's constitutes the framework of space-charge-limited conduction (SCLC) (Lim et al., 2016).

#### **1.1.4 Nanomaterials**

Nanomaterials are defined as any particulate with a diameter of less than 100nm. Ag nanoparticles is chosen from all the other metallic nanoparticles due to their high work functionality and chemical stability. This allows them to be useful for charge trapping in non-volatile memory applications. With the supporting evidence from previous studies, silver nanoparticles are chosen as the inclusion in the polymannose thin film. Silver nanoparticles as raw material are mostly available in 99.95% purity in powder form due to their easily oxidizing properties. Usually, the raw material will be coated with a layer of oxide. This oxide layer formed is considered one of the intrinsic properties in this study since the presence of oxygen vacancies also plays a vital role in the device's resistive switching behavior.

#### **1.2 Problem statement**

Polymannose is being studied as a sustainable bio-organic material to be used as the resistive film in memristors. It is reported by Tayeb et al. (2020) that polymannose being dried at 160°C for 7 hours showed excellent resistive behavior with 12 multistate at a low form voltage. However, this study only provides insight into the organic polymannose application without any alteration of the material. A study on metallic nanoparticles inclusion in the film should be carried out to explore the full potential of the polymannose film for future ReRAM development. A better understanding of metallic nanoparticles on the effect of the resistive switching mechanism and behavior of the polymannose film can be examined through this work. In this paper, different concentrations of Silver Oxide nanoparticles are used as the parameter to determine the optimum metallic nanoparticles concentration for the ideal resistive switching behavior. The addition of metallic nanoparticles is expected to improve the resistive switching behavior.

### **1.3 Objectives**

- i. To fabricate a resistive switching device using polymannose thin film with the addition of silver oxide nanoparticles at different concentrations.
- ii. To study the effect of silver oxide nanoparticles incorporation on the resistive switching properties of polymannose layer as an insulator film in the device.

### **1.4 Research approach**

Polymannose thin film has shown remarkable properties that are important in Resistive random-access memory (ReRAM) and multistate switching memory applications. Some of these properties include a high ON/OFF ratio, relatively low READ voltage, high endurance cycle and long retention time. For these reasons, polymannose thin film is said to be a potential bio-organic material in producing the next-generation artificial synapses. However, the resistive switching behavior of polymannose thin film loaded with metallic nanoparticles has not yet been explored. Therefore, this study aims to examine the effect of different concentrations of silver oxide nanoparticles incorporated in the polymannose thin film. With reference to Tayeb et al. (2020), the device can be fabricated with D-mannose powder with ethanol as precursor with different concentration of silver oxide nanoparticles added which are 0.5 wt%, 1.0 wt%, 2.0 wt% and 10 wt%. The mixture can then be drop casted on the film and left dried for 7 hours at 160°C. Characterization of material regarding its surface tension, thickness, morphology, and topology can be carried out using High Resolution Transmission Electron Microscopy – Energy Dispersive X-ray (HRTEM-EDX), Atomic Force Microscopy (AFM), Fourier Transform Infrared Spectroscopy (FTIR), Scanning Electron Microscopy (SEM), and Goniometer. It is commonly believed that metallic nanoparticles serve as a

metallic source for the redox process in the electrochemical mechanism and the addition of trapping and detrapping site in the electric mechanism. Thus, it is hypothesized that the addition of these metallic nanoparticles can reduce the resistivity of the thin film and enhance the memristive characteristics. The resistive switching properties of the device can be characterized by using Semiconductor Parameter Analyzer. The knowledge gained from this work will be highly beneficial for future development of the ReRAM.



## **CHAPTER 2**

### **LITERATURE REVIEW**

#### **2.1 Introduction**

This chapter reviews the resistive switching behavior in non-volatile memory devices and bio-organic materials applied in ReRAM. This development is systematically reviewed according to the timeline and material used as the resistive thin film in the non-volatile memory device. A more in-depth review is done specifically on the aloe vera application in bio-organic based memory devices. A detailed discussion on the performance of the device especially its resistive switching behavior which includes the factors, constant and manipulated variables, and characterization method is presented.

#### **2.2 Memristor**

##### **2.2.1 Introduction to resistive random-access memory (ReRAM)**

In the era dominated by the Internet of Things (IoT) with the rapid emergence of artificial intelligence, the von Neumann architecture is struggling to stay at its peak performance. Energy consumption is the major holdback of the Von Neumann architecture due to the presence of a gap between the central processing unit and memory unit, restricting data process and storage to be carried out simultaneously. Additionally, electronic chips designers had tried to improve chip performance by increasing the number of transistors, however, this causes another problem to arise. Heat generation from many transistors has impacted the function of the device. To overcome this, a highly efficient and high-density hybrid circuit incorporated with memristors is proposed. Memristors with fast writing and rewriting capabilities, non-volatility, and brain-like highly efficient systems are quickly replacing flash memory and DRAM (Dynamic random-access memory).