SCHOOL OF MATERIALS AND MINERAL RESOURCES ENGINEERING

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FABRICATION OF POLYSILOXANE BASED STRETCHABLE INK FILLED WITH SILVER NANOPARTICLES

By

Siew Tuck Seong

Supervisor: Prof. Dr. Zulkifli Ahmad

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DECLARATION

I hereby declare that I have conducted, completed the research work and written the dissertation entitled: "**Fabrication of Polysiloxane Based Stretchable Ink Filled with Silver Nanoparticles**". I also declare that it has not been previously submitted for the award of any degree or diploma or other similar title of this for any other examining body or university.

Name of student	: Siew Tuck Seong	Signature:
Date	: 25 June 2018	

Witness by

Supervisor	: Prof. Dr. Zulkifli Ahmad	Signature:
Date	: 25 June 2018	

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Abbreviations	Description
AgNPs	Silver Nanoparticles
AgNWs	Silver Nanowires
CuNWs	Cooper Nanowires
CNTs	Carbon Nanotubes
MWNTs	Multi-walled Carbon Nanotubes
FTIR	Fourier Transform InfraRed Spectroscopy
DSC	Differential Scanning Calorimetry
SEM	Scanning Electron Microscope
OM	Optical Microscope

FABRIKASI DAKWAT YANG BOLEH REGANG BERDASARKAN SUBSTRAT POLISILOKSANA DENGAN NANOPARTIKEL PERAK

ABSTRAK

Dakwat konduktif boleh regang adalah salah satu teknologi yang sedang dibangunkan untuk dekad ini. Dalam kajian ini, gabungan komposit konduktif disediakan dengan menggunakan polidimetilsiloksana (PDMS) sebagai substrat dan nanopartikel perak (AgNPs) sebagai bahan konduktif. Bahan-bahan yang digunakan untuk penyediaan substrat adalah polidimetilsiloksana hidroksida (PDMS-OH), wasap silika, toluene, dibutyltin dilaurat dan 3-glycidoxypropyl-3 trimethoxysilane. Dakwat konduktif perak dicetak pada substrat PDMS dan matang pada 80 ℃ selama 16 jam. Pelbagai ujian telah dijalankan untuk mengkaji sifat-sifat elektrik, mekanik dan kimia komposit PDMS-Ag. Sampel PDMS-Ag menunjukkan rintangan elektrik rendah yang mencerminkan kekonduksian yang baik dari sampel apabila regangan. Selain itu, sampel PDMS-Ag menunjukkan rintangan elektrik rendah yang munasabah selepas pelbagai kitaran 50% regangan terikan. Selain itu, komposit PDMS-Ag juga memaparkan pengikatan antaramuka yang baik antara matriks PDMS dan dakwat konduktif Ag melalui ujian lekatan.

FABRICATION OF POLYSILOXANE BASED STRETCHABLE INK FILLED WITH SILVER NANOPARTICLES

ABSTRACT

Stretchable conductive ink is one of the developing technology for the recent decades. In this research, stretchable conductive composite is prepared by using polydimethylsiloxane (PDMS) as substrate and silver nanoparticles (AgNPs) as the conductive material. The ingredients used for the substrate preparation are polydimethylsiloxane-hydroxide (PDMS-OH), fumed silica, toluene, dibutyltin dilaurate and 3-glycidoxypropyl-3 trimethoxysilane. The silver conductive ink was printed on the PDMS substrate and cured at 80 ℃ for 16 hours. Various tests have been conducted to study and investigate the electrical, mechanical and chemical properties of the PDMS-Ag composite. The PDMS-Ag sample shows low electrical resistance which reflects to good conductivity of the sample upon stretching. Furthermore, the PDMS-Ag sample shows reasonable low electrical resistance after various cycles of 50% strain stretching. Besides, the PDMS-Ag composite also displays good interfacial bonding between PDMS matrix and Ag conductive ink through the adhesion test.

CHAPTER 1

INTRODUCTION

1.1 Research Background

Most of the electronics are known as bulky and rigid components since they are manufactured on rigid substrates such as glass and/or silicon. However, the future of the electronics may be approaching in the direction of electronics that manufactured on polymeric foils, subsequently going to be flexible and even stretchable. The general developing trend of electronics is heading in the way of development flexible and stretchable electronics (Dang *et al.*, 2017). The main reason behind the approach of electronics towards this trend is the limitation of the traditional electronics which has the properties of brittle properties that limit the ability of the circuit to be stretched or bended.

The development of stretchable electronics would bring a positive impact in the development of electronic skin (E-skin). E-skin is a wearable electronic that can be very useful in many applications especially in medical field. It can aid in monitoring individual's health condition such as pulse rate, blood pressure and other vital signals. Those data can be recorded and assist doctors to make a more precise diagnosis on patients E-skin is an electronic device that imitated the real human skin which can be flexed, bended and stretched without incurring any physical damage (Hammock *et al.*, 2013).

In the development of stretchable electronics, the main challenge to make silicon - based structures stretchable since the brittleness of silicon makes it almost impossible to be stretched. Many researches bypassed this difficulty by using stretchable interconnects (Someya, 2013). A successful strategy to stretchable electronics uses post-buckling of stiff, inorganic films on compliant, polymeric substrates. According to Amjadi et al.

(2014), they found that the fabrication of conductive silver nanowires thin film embedded between two layers of Polydimethylsiloxane (PDMS) can achieve a sandwich structure with excellent flexibility, stretchability and bendability which can also conduct electric. Besides, using conductive ink on the polymeric substrate is another way of fabrication of stretchable electronics. Conductive ink is a type of ink that infused the conductive material into the ink. The commonly used conductive material would be silver. It is because silver has a high conductivity and almost no change in conductivity as the silver particles. Conductive ink has the advantage to be fabricated into wide range of sizes and shapes. The fabrication of the silver conductive ink on PDMS substrate is shown in Figure 1.1.

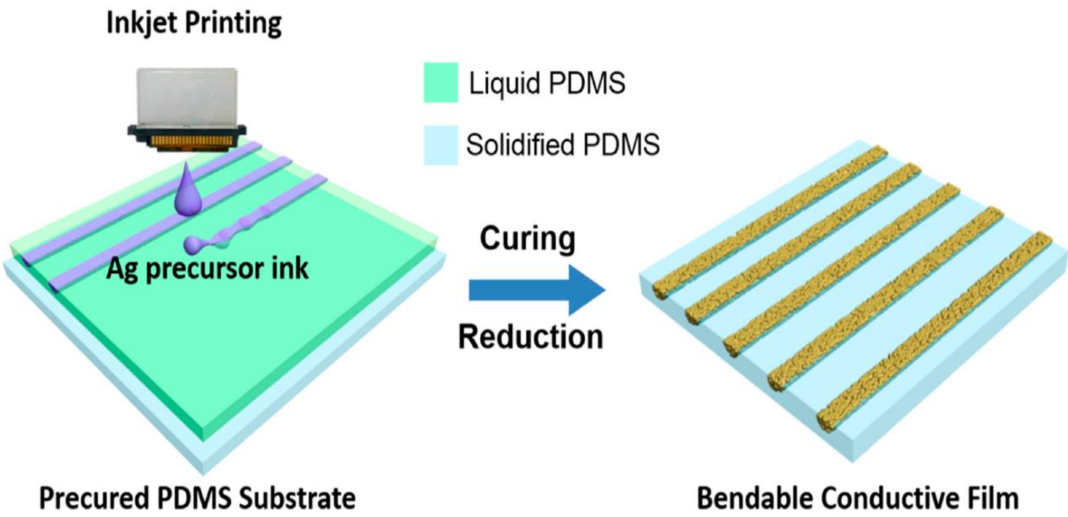

Figure 1.1 Fabrication of conductive film by using silver conductive ink on PDMS substrate (Sun *et al.*, 2016).

PDMS is a type of silicone rubber where the chemical structure is shown in Figure 1.2. It has become the most commonly used elastomer in rapid prototyping of microfluidic devices because of its simple fabrication procedure through casting and curing onto a microscale mold and strong sealing to a wide variety of materials (Galindo-Rosales, 2017). It offers many advantages such as good flexibility, temperature stability from −50 ◦C to +200 ◦C, chemical inertness, low cost and simple fabrication (Galindo-Rosales, 2017). This silicone rubber was initially developed as high-voltage outdoor insulator material with low surface tension (hydrophobicity). PDMS also has favourable optical properties including transparency above ∼230 nm and very low auto fluorescence over a wide range of wavelengths compared to other traditional silicon chip materials. Furthermore, PDMS is permeable to gasses, impermeable to water and non-toxic to cells, making it suitable for biomedical application, especially in the development of E-skin.

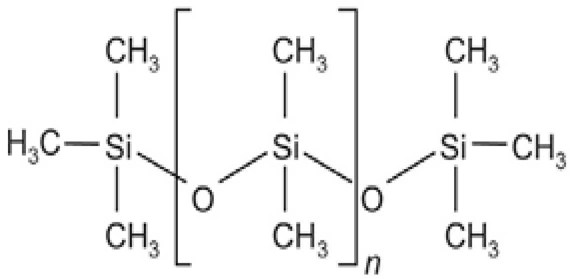

Figure 1.2 Structural formulation of PDMS oligomer (Berean *et al.*, 2014).

Silver ink is one of the mostly used conductive materials in the fabrication of stretchable electronics thanks to its relatively high and consistent electrical conductivity compared to other conductive materials. Besides, silver ink also have strong anti-oxidation properties which can prevent growth of bacteria and fungi which makes it perfect in the stretchable electronics in biomedical field (L. Y. Xu *et al.*, 2014).

The adhesion between the silver ink and PDMS substrate raised concern to researchers. These two materials are ideally suitable for fabricated stretchable electronics, especially in the biomedical field. However, the hydrophobicity of PDMS hinders wetting of the PDMS surface by silver nanowires (AgNWs), resulting in poor mechanical stability of the fabricated electrodes (Kim *et al.*, 2015). Therefore, it is very crucial to ensure that there is good interfacial interaction and adhesion between silver ink and PDMS, in order to fabricate an electrode with consistent conductivity to endure repeating stretching and releasing.

1.2 Problem Statement

The challenge of this research is to improve the consistency of electrical conductivity property of the stretchable conductive ink. The issue of consistency is very much crucial when working materials at nanoscale level. Nanoscale particles has very much variable shape and sizes which is subject to several orientation and stacking depending on slightest changes in processing techniques. The fabricated stretchable conductive ink is required to have excellent electrical conductivity as the ink is under mechanical deformation such as stretching. In other words, this project aims to produce an excellent electrical conductivity of stretchable conductive ink under certain strain rate.

Besides, the adhesion of the conductive silver nanoparticles (AgNPs) with the polysiloxane based substrate, polydimethylsiloxane (PDMS) is a challenge for this research. The conductive ink is needed to have good adhesion between the conductive material and the polymeric substrate. The crucial requirement is that both the ink and the substrate should have a comparable elastic modulus such that during stretching, they will strain at similar amount without affecting any local rupture. Further both the ink matrix and the substrate is made up of similar polymeric material which ensure a good interfacial interaction hence improve adhesion.

These are the concerns and challenges of this research project. The outcome of this research may be contribute in the technology of the stretchable electronic especially in the e-skin industry.

1.3 Objectives

The main goal of this research project is to prepare a polysiloxane based stretchable ink filled with AgNPs. The findings in this research would be essential in the development of stretchable electronics. To achieve this goal, there are two main objectives:

1. To design stretchable conductive polymer ink formulation based on polysiloxane and silver nanoparticles.

2. To study the electrical properties of the polysiloxane based stretchable ink filled with silver nanoparticles under static and dynamic stretching condition.

CHAPTER 2

LITERATURE REVIEW

2.1 Stretchable Electronics Overview

There are many factors have to be considered in fabricating an excellent performance stretchable electronics. Researchers went in several direction to develop functional and consistent stretchable electronics, including stretchable conductors, stretchable semiconductor, stretchable insulator, stretchable electronic devices with intrinsically stretchable components and etc. According to Trung & Lee (2017), the fabrication of stretchable electronics can be categorised into two different approaches: stretchable components that is obtained by geometric engineering and stretchable components with intrinsic stretchability.

In the first approach, the beauty of nanoelectromechanical system has provided a resolution for the traditional rigid Si electronic. Generally, the traditional Si electronics are barely able to be twisted and stretched. When a 100nm thick of Si film is integrated with a 20 µm thick elastic polymer substrate. The Si film is capable to be bended in radius of curvature 10mm (Wang, 2016). However, this mechanical flexibility does not cope to meet up more critical mechanical deformation that come with the formation of massive stretchability. Therefore, the brilliant of structural engineering introduces the modification of the geometric structure of non-stretchable material into wavy (Xu *et al.*, 2012), buckled (Song *et al.*, 2015), wrinkled (Oyewole *et al.*, 2015), serpentine (Xu *et al.*, 2013), island (Song *et al.*, 2015) and net configurations(Rogers, Someya and Huang, 2010). According to Rogers, Someya and Huang (2010), this geometrical approach enable the rigid silicon to be stretched with a strain range of 10 to 20% which is 10 to 20 times as large as the intrinsic fracture limits of the silicon. However, Trung & Lee (2017), think that this fabrication process of stretchable electronics is very complicated, not cost efficient and many uncontrollable factors which is not so ideal in commercialise the stretchable electronics.

As the alternative, there is an approach in developing the stretchable electronics based on stretchable components with intrinsic stretchability as shown in Figure 2.1 (Trung and Lee, 2017). This involve the electronic components themselves which are stretchable eg. transistor, capacitor and diode. This approach has the advantages of simple fabrication, high yield, and low cost. However, there is a major drawback of this approach which is the difficulties in obtaining all the stretchable components; conductor, insulator and semiconductor materials.

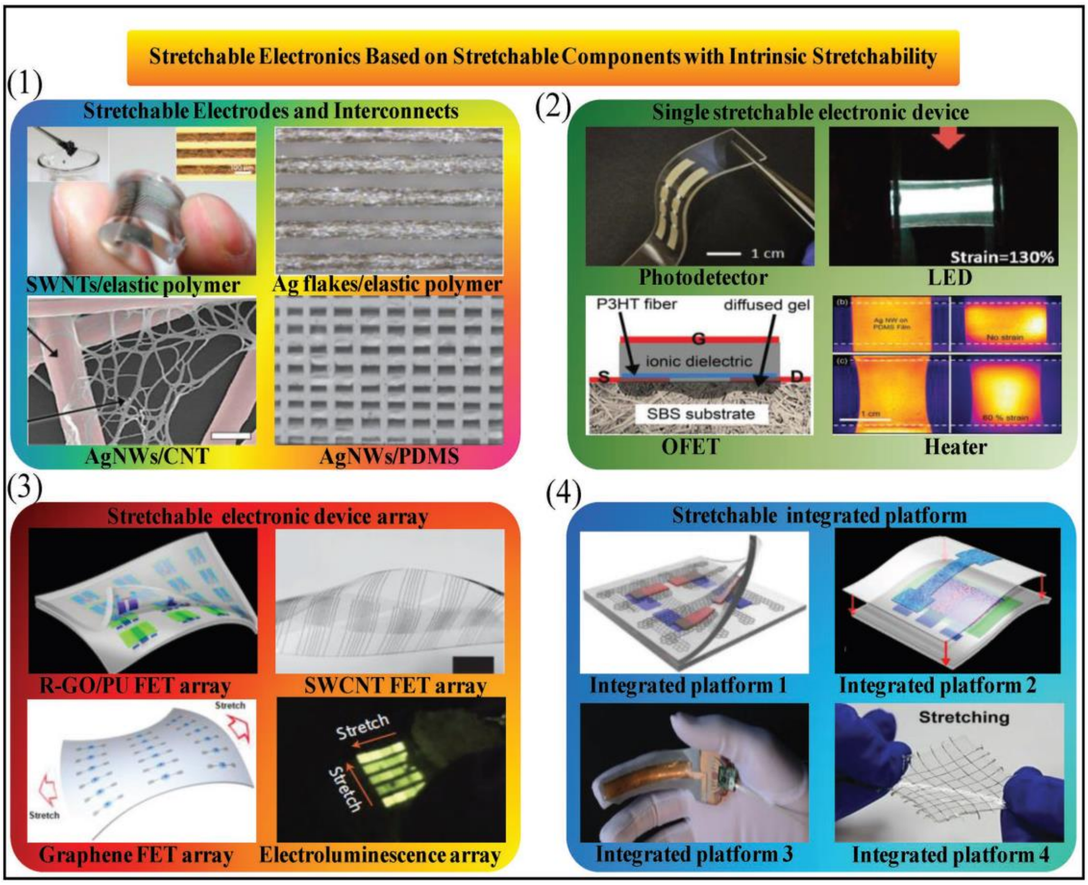

Figure 2.1 Stretchable electronics based on intrinsically stretchable components.(Trung and Lee, 2017)

2.2 Stretchable Electronic Applications

Stretchable electronics can be used and applied in varies field of applications. There are two major application fields for stretchable electronic system. They are electronics close to human body and the electronics that attached to three dimensionally curved surfaces (Loher, Seckel and Ostmann, 2010).

The application of stretchable electronic that close to human body can be further divided into medical and textile electronics. The major aim of medical stretchable electronic is the integration of sensor and actuator for medical monitoring and controlling.

From the inspiration of human skin, the artificial skin (as known as E-skin) has developed to imitate human skin that provide tactile information which can be used to evaluated the parameters for object handling (Yogeswaran *et al.*, 2015). Furthermore, this artificial skin also provide the sensation information such as surface compliance and hardness of object which is able to help in the development in the field of prosthetic limbs.

Besides, in field of actuator development, Kang and Pak (2017) reported that the arrhythmias disease is facing a challenge to be diagnose and treated due to the complex component structures and 3D regions of heart. Therefore, a flexible and stretchable balloon like catheter was designed with multiple sensors surrounding the outside surface as shown in Figure 2.2. These sensors enable the catheter to obtain temperature, flow characteristics, tactile, optical, and electrophysiological data from the tissue-balloon interface.

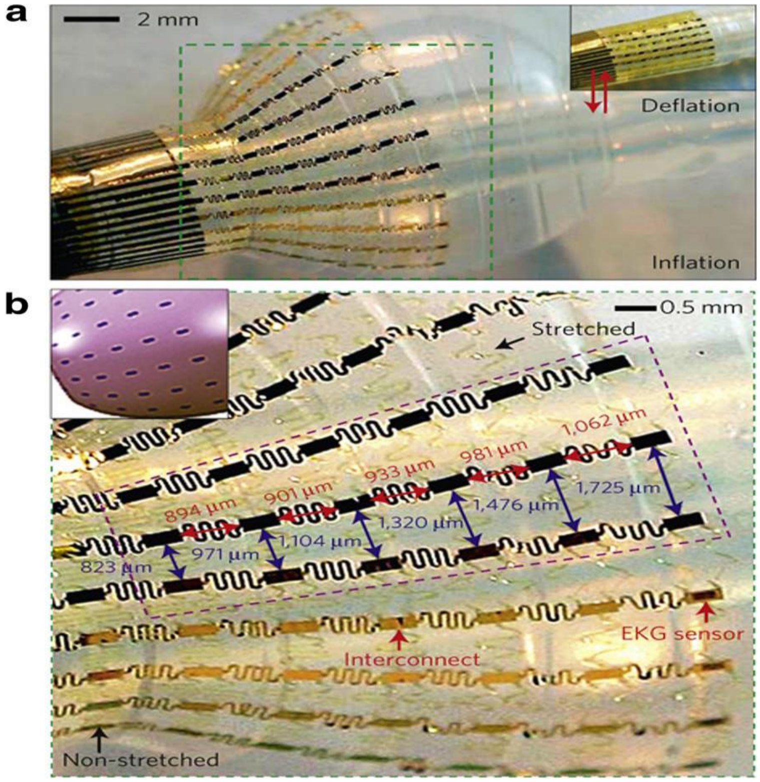

Figure 2.2 : Image of inflatable catheter surrounded with multiples sensors(Kang and Pak, 2017).

Commonly, the implantable devices usually cause a damage to the organs or acts as a source of infection by the tissue immune reaction. Therefore, the implantable devices required to be attached closely to the organs, especially heart, and cause no immune reaction from the organ. To implant sensors or actuators tightly on heart surface is very challenging. It is because heart expands and contracts as it beats in rapid rate which lead to detachment of sensors from the surface. Bowden et al. (1998), reported that PDMS is one of the polymers that can deform up to 200% of strain under heating and cooling condition. Furthermore, the PDMS is an organic polymer that will not induce infection by the tissue immune reaction (Bowden *et al.*, 1998). Hence, the sensors can be fabricated on PDMS substrate and functional under rapid rate of expanding and contracting without being detached from the heart as shown in Figure 2.3.

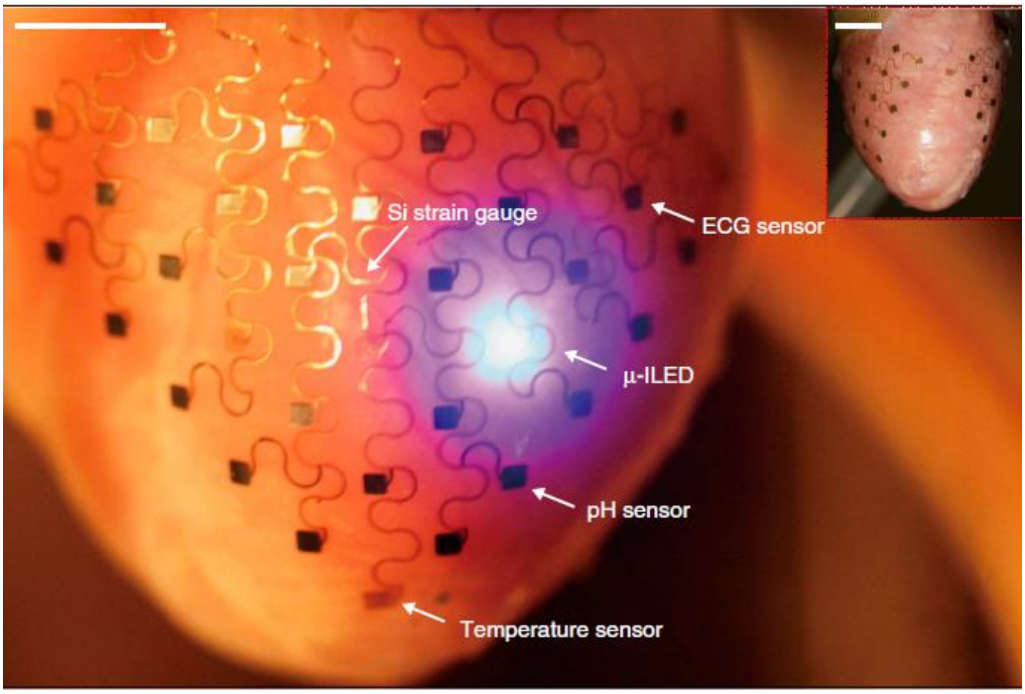

Figure 2.3 : Image of the stretchable electronics devices with multiple type of sensors integrated on a Langendorff-perfused rabbit heart (L. Xu *et al.*, 2014).

As shown in Figure 2.4, it is a device that function not only as a sensor but also an actuator. This device consists of an array of 8 electrodes around the heart circumference and capable to deliver spatially and temporality programmed electrical stimulation onto the heart outside membrane (Xu *et al.*, 2015).

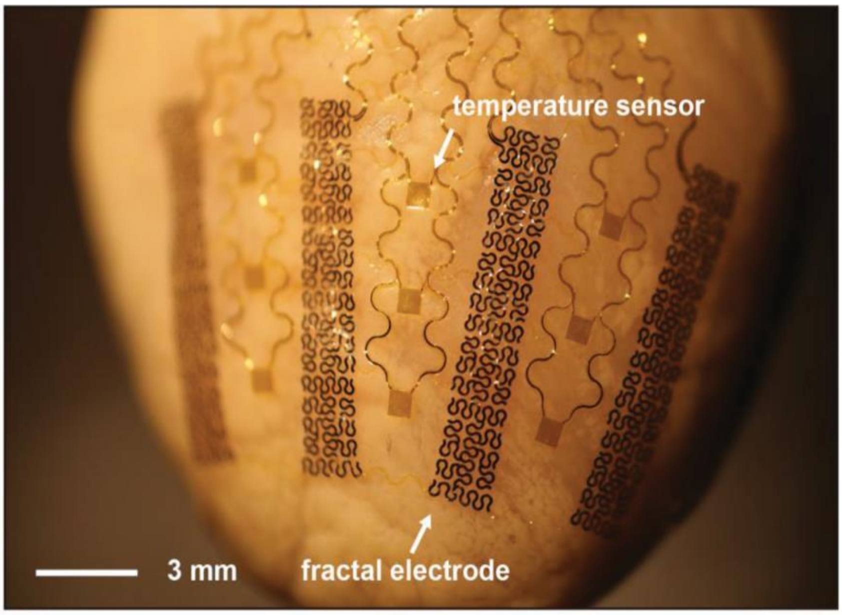

Figure 2.4: Image of a representative device integrated on a rabbit heart(Xu *et al.*, 2015).

Next, there is another major application of stretchable electronics which is in the field of textile. The stretchable electronics provide electronic support for technical textile usage and wearable computing (Loher, Seckel and Ostmann, 2010). The stretchable electronic textiles are expected to be capable to withstand cyclic stretch of 3-5% of strain and robust against moisture due to human body liquids or repetition of washing.

According to Oh, Park and Kim (2003), they reported that a stretchable conductive fabric for electrotherapy. The electrotherapy uses electric pulses to in physiotherapy and rehabilitation to reduce pain, enhance healing and improve patient mobility. The stretchable fabric displays well maintained conductivity even at 40% of extension. Therefore, it enhances the properties of the stretchable fabric to maintain consistent electrical properties to perform electrotherapy despite the fabric is subjected to various movement of human body (Oh, Park and Kim, 2003).

Besides, Dong et al. (2017) reported a highly stretchable and washable all-yarn-based self-charging knitting power textile. This textile enables biomechanical energy harvesting and simultaneously energy storing by hybridizing the triboelectrical nanogenerator and supercapacitor in one fabric. This self-charging knitting power textile can continually operate a temperature-humidity meter or a calculator by hand tapping it (Dong *et al.*, 2017). In addition, this self-charging power textile also possess high elasticity, flexibility and stretchability. This self-powering stretchable and wearable electronic displays extensive possibilities for the stretchable electronics and the sustainable energy system.

2.3 Stretchable Conductors

In the fabrication of stretchable electronics, the selection of stretchable conductors is the one of the major factor to be put into consideration. In section 2.1, it is been discussed that there are two different approaches in the fabrication of stretchable electronics. There are varies of approaches in fabricating stretchable electronics based on the intrinsic properties of the stretchable components. Without using the complicated geometric engineering process, generally the stretchable conductors are fabricated from Ag nanowires (AgNW), Cu nanowires (CuNWs), carbon nanotubes (CNTs) and conductive polymers (Trung and Lee, 2017). However, according to Yuan et al. (2018), the metal nanowires are more desirable conductors in the fabrication of stretchable electronic. It is because the carbon nanomaterials do not have excellent and stable electrical conductivity during straining process which limited the usage in some of the stretchable electronics applications.

2.3.1 Copper Nanowires (CuNWs)

There are three metal in group 11 of the periodic table which are commonly known as great electrical conductors; they are copper (Cu), silver (Ag) and gold (Au). These three metal conductors had made a lot of contribution in the electrical world due to their intrinsic properties, an excellent conductor of electricity. Copper (Cu) is a material known as one of the common metal electrical conductor. It is extensively used in many electrical applications, especially wire and cables.

AgNWs is one of conducting materials many researchers are using to develop stretchable conductors, Copper Nanowires (CuNWs) are considered as well as potential material for stretchable conductor or the alternative material of AgNWs. It is because Cu is more abundant and almost hundred times cheaper than Ag, while the electrical conductivity is still comparable with these metals with highest conductivity (Han *et al.*, 2014).

Cheng et al. (2014) fabricated a CuNWs with the elastic poly(acrylate) matrix transparent conductive film that shown 91.5% of transparency with 220 Ω/□. However, in term of conductivity of this film in stretching condition, its resistance had increased double when the sample reached its rupture strain which is around 15% (Cheng *et al.*, 2014). However, there is another method that prepared the CuNWs by using plasmonic laser nanowelding method. Plasmonic laser nanowelding is a method that is capable to form a CuNW percolation network by welding up the nanowires junctions together to increase the contact area which can instantaneously reduce the electrical resistivity to 20 Ω/□ (Han *et al.*, 2014). This method is used to minimize the oxidation that occur on the CuNWs that processed by the conventional thermal treatment under ambient condition.

2.3.2 Carbon Nanotubes (CNTs)

Carbon nanotube (CNT) is a form of carbon, with nanometer-sized diameter and micrometer-sized length. CNTs is a member of fullerene structural family which is a type of carbon allotropes. CNT is a cylindrical shaped with opened ends which is a most distinctive difference between the fullerene structural family (Zaporotskova *et al.*, 2016).

CNTs have high aspect ratio as well as high thermal stability, mechanic robustness and most importantly, CNTs also possess of good electrical conductivity properties which made them can be considered as a potential material for stretchable conductors (Zhang *et al.*, 2010). Zhang et al. (2010) reported that they fabricated a transparent stretchable conductors that the electrical conductivity of the film can be maintained in stable condition after repetition of stretching. It is a conductive film that is based on aligned CNT ribbons and PDMS elastic matrix. The electrical resistance of this film increased in 1.5 times when the film is under 220% of strain. Interestingly, the resistance of this film is nearly unchanged with the condition of 100% strain and 30 stretching cycles. Besides, Lee et al. (2014) also reported a hierarchical multiscale AgNW/CNT hybrid nanocomposite that is highly stretchable (>460% of strain), remarkable mechanical properties including over 10000 times of bending, over 540 ° of twisting and complete folding. Furthermore, AgNW/CNT hybrid nanocomposite also come with high transparency (80-93%). This excellent mechanical realibility is due to the excellent mechanical properties of CNT meanwhile AgNW contributed to the conductivity of the film (Lee *et al.*, 2014). Same as AgNWs, CNTs are still expensive to be produced (Aqel *et al.*, 2012). Furthermore, its electrical conductivity is not as comparable as the traditional metal conductors.

2.3.3 Silver Nanowires (AgNWs)

Among all the potential metal nanowires, AgNWs have shown the highest electrical conductivity among metallic nanowires. The one-dimensional wire structure with a superior aspect ratio is better to form electrical percolation network than particles or flakes. Therefore, AgNWs can maintain good electrical conductivity under deformation, especially during stretching (Yuan *et al.*, 2018). Therefore, it is a suitable material as the promising candidate in the fabrication of stretchable electronics.

Akter and Kim (2012) fabricated a transparent and stretchable conductive thin film based on AgNWs. This conductive thin film had successfully maintain its low resistance at the value of 35Ω/□ with the 15-20% of strain. It is suggested that the conductivity of the thin film can be maintained at higher strain if more AgNWs are deposited on the film (Akter and Kim, 2012). It is because according to the percolation theory, when the conductive filler concentration is above the percolation threshold, the conductive filler may form a network that allow direct current flow through it. However, when the amount of AgNWs increased the Young's Modulus of the film may increase at the same time which may reduce the stretchability and the optical transmittance of the film(Trung and Lee, 2017). Therefore, it is a massive challenge to manipulate the amount of the conductive filler that can produce a stretchable conductor that capable to maintain high conductivity under high strain condition meanwhile also produce excellent optical properties.

2.3.4 Silver NanoParticles (AgNPs) Conductive Ink

Previously it was discussed that silver is one of the common used conductive material in the electronic field. Furthermore, the Ag has the outstanding conductivity and excellent oxidation resistance among these three notable conductive metals. In addition, Ag also have a remarkable antibacterial properties which can avoid the growth of bacteria and fungi on the inks (Rajan *et al.*, 2016). This is a very important characteristic, especially for the development of stretchable electronics in the biomedical field, such as in the personal health care monitoring and medical prosthetics which are strictly demanding for the hygiene of the electronic devices (Yogeswaran *et al.*, 2015).

The morphology of the selected metal particles and the ability of binder to carry these particles during printing and curing is very crucial factor in the fabrication of stretchable ink (Mohammed, 2017). Therefore, Rajan et al. (2016) suggested and fabricated silver nanoparticles (AgNPs) conductive ink due to AgNPs are able to be connected at nano-scale level and reducing surface tension and optimizing ionic forces that give stability to a suspension in the form of ink. Besides, nanoparticles also have the electron resonances that allows interactions with electromagnetic field and enabling sintering at low temperature. However, the most important feature of AgNPs compared to AgNWs is the shape and dimension of AgNPs are easier to be control and less expensive to be processed. Therefore, more and more researchers are heading in the development of conductive silver ink.

2.4 Substrate/Matrix for Stretchable Electronics

Generally, the traditional conductive material including for the nano-sized conductive material intrinsically do not have the elastic properties that can be recovered into original length after repetition of stretching cycles. Therefore, researchers had another way out which is fabricated the conductive composite based on elastic polymer substrate. The reason is due to elastomers are viscoelastic material intrinsically can be expand reversibility in many times into their original length. However, the conventional circuit board are extremely brittle and not suitable in the development of stretchable electronics (Someya, 2013).

Elastomer plays an important role in the fabrication of stretchable electronics thanks to its low elastic modulus and high yield strain. This remarkable properties suits perfectly for the stretchable electronics (e-skin) to imitate real human skin which can be bended, twisted and stretched in multiple cycles without incurring any physical damage

(Hammock *et al.*, 2013). With the presence of elastic polymer, it allows researchers to focus more on the stretchable conductive materials.

2.4.1 Polyurethane (PU)

The development of elastic PU began because during World War II the sources of natural rubber was expensive and difficult to be obtained. Therefore, researchers develop PU as an alternative. Hence, PU shows some many remarkable properties as an elastomer; high abrasion and tear strength, better resistance to oxygen aging while displaying good flexibility and elasticity (Prisacariu, 2011).

Polyurethane (PU) is a polymer with the presence of urethane bonds. The urethane group commonly formed by the reaction between isocyanate and hydroxyl groups. The presence of diisocyanate and marco-diol are the key components that make PU so special. The diisocyanate and polyol are termed as the hard segment and soft segment of the structure. The hard segment usually is a rigid aromatic molecule with Tg above ambient temperature meanwhile Tg of the soft segment is usually lower than the ambient temperature (Hepburn, 1992).

There are researches on development of stretchable electronics based on PU, Noh (2016) reported a research that prepared elastomeric polyurethane-polypyrrole composite foams with the conductivity 10^{-5} S/cm and an elongation at break of 160%. It has reflected that PU foam is not suitable in the development of stretchable electronics due to the low conductivity of the composite (Noh, 2016).

Furthermore, Shang, Zeng and Tao (2011) developed an elastic conductive nanocomposite made with multiwall carbon nanotubes (MWNTs) and PU. It is reported that this MWNTs/PU nanocomposite maintained its conductivity under 100% of strain and certainly the nanocomposite can be stretched more than 100%. In addition, this nanocomposite shows a promising durability by showing that it is still able to conduct electricity after 100 cycles of stretching/relaxing mechanism (Shang, Zeng and Tao, 2011). However, there is major drawback that raise the concern of the researchers to use PU as the stretchable substrate in the wearable stretchable electronics. It is because the isocyanate group in the PU structure will lead to health effect when human exposed under short/long term. Even traces of isocyanate vapour may cause bronchial troubles, especially for the people with asthmatic issue (I. R. Clemitson, 2015). PU is a safe material when it is well-manufactured. However, some of researchers may look for alternative material to provide stretching properties instead of PU.

2.4.2 Silicone Rubber – Polydimethylsiloxane (PDMS)

Polydimethylsiloxane (PDMS) is the basic skeleton of the types of silicone rubber available in the market. Generally, PDMS is categorized into two types; heat-vulcanized type and room-temperature-vulcanized type. The heat-vulcanized type silicon rubber is characterized by extremely high molecular weight (300,000-700,000), meanwhile the room-temperature-vulcanized type of silicon rubber is relatively lower molecular weight (10,000-100,000). This relatively lower molecular weight suits the processing method such as dipping and spreading (Hecht, 1968). Therefore, PDMS with ranges of molecular weight can provide specific mechanical properties and viscosity for particular processing method and application purpose. Besides, PDMS also has the properties of high electrical resistance (2.9×10^{14} Ohm.cm), low glass transition temperature (-125 ℃), large thermal coefficient of expansion ($4.8 \times 10^{-4} K^{-1}$) and high flexibility (with Young's module of 1MPa) (Zhao and Huang, 2017).

Many researchers have chosen PDMS as the stretchable substrate in the development of stretchable electronics, especially in the development of biomedical and wearable electronics. Several fabrication methods by using the combination of varies conductive material with PDMS have been reported (Trung and Lee, 2017). As per Trung and Lee (2017) reported, there is a composite of AgNWs pattern embedded into PDMS showed a low sheet resistance (9 Ohm/ sq), high stretchability (50%), good chemical stability and high uniformity. Besides, Yoon and Khang (2016) fabricated a transparent electrodes welded and patterned by AgNWs network on the patterned PDMS stamp. This transparent electrodes can be bended up to 10% and stretched up to 40% without losing its electrical properties. In addition, this electrodes have a high optical transparency around 85-90% (Yoon and Khang, 2016). Furthermore, there is another elastic conductor made by Single Walled CNTs embedded in PDMS that showed remarkable results. This composite can be stretching 70-100% uniaxial and biaxial without mechanical or electrical damage.

However, there is a raised concern for using PDMS as interconnect of the conductive material. It is because PDMS is made out of hydrophobic structure and it is chemically stable yet inert material. Therefore, the conductive materials would be having difficulty to have a good adhesion on PDMS. To improve the adhesion between the PDMS and the conductive material, there are several methods that have be done to solve this problem.

The first method is reported by Amjadi et al. (2014), they used sandwich structure processing method to embed AgNWs in the PDMS as shown in Figure 2.5.

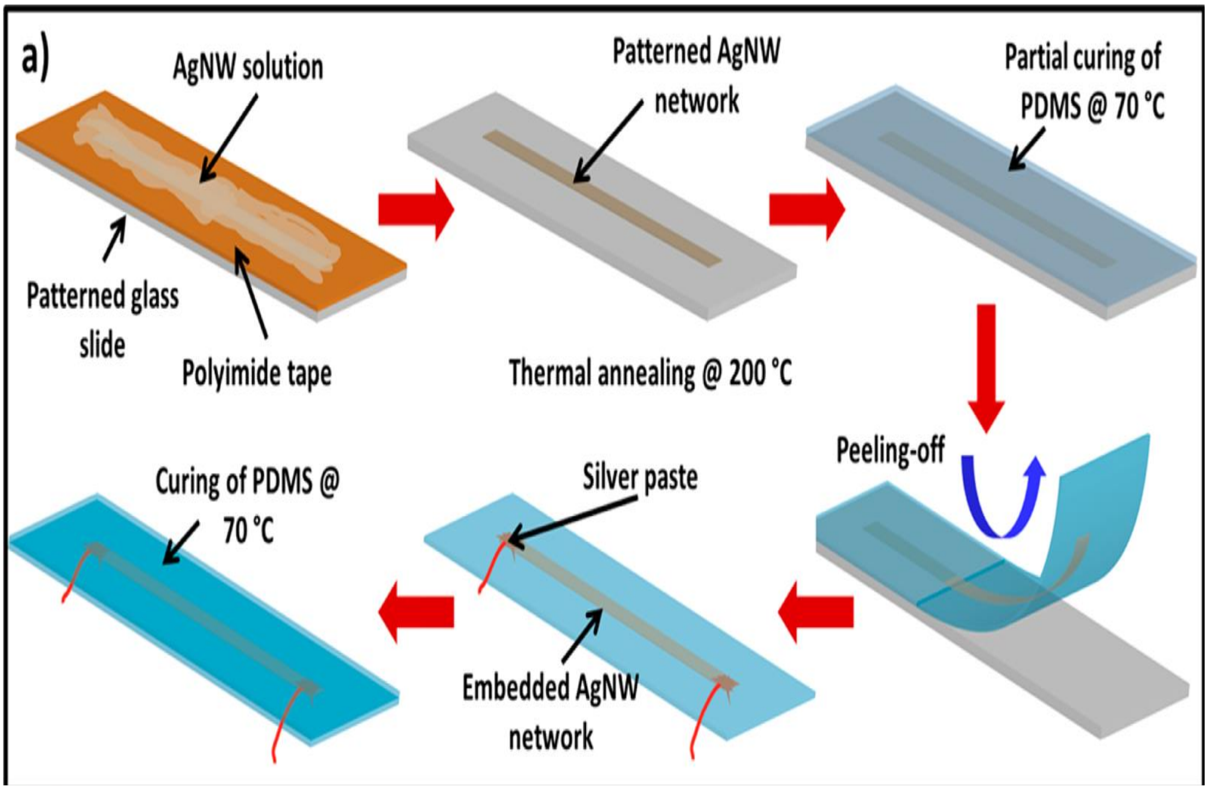

Figure 2.5 Fabrication process of the sandwich-structure PDMS/AgNW/PDMS nanocomposite strain sensor (Amjadi *et al.*, 2014).

The following approach to fabricate a good adhesion between the PDMS and the conductive material, Yuan et al. (2018) used the screen-printing method to fabricate the stretchable circuit. Figure 2.6 illustrates the fabrication process of patterned AgNWs/PDMS stretchable electrodes. Firstly, the prepared Ag NWs paste was screen printed on glass substrate to pattern conductive circuits, and then the printed Ag NWs patterns were dried in a vacuum oven to remove solvent and to form a uniform and conductive film of Ag NWs network. Next, freshly mixed PDMS liquid was casted on top of the printed Ag NWs patterns and to peel off the substrate after curing at 80 ℃ for 2 h. In this way, the randomly oriented Ag NWs were buried in PDMS to form a conductive and stretchable circuit. Yet, there is no certain resolution to acknowledge the adhesion of PDMS with conductive material, especially after cycles of stretching mechanism. Furthermore, Akter and Kim (2012) used surface treatment process on PDMS with dopamine to improve the adhesion of PDMS with AgNWs. By using dopamine, there is no significant amount of AgNWs peel off after the adhesion test which is performed by peeling off 3M scotch tape adhere on the AgNWs/PDMS as shown in Figure 2.7. In addition, the AgNWs/PDMS films still maintain its electrical conductivity after the adhesion test.

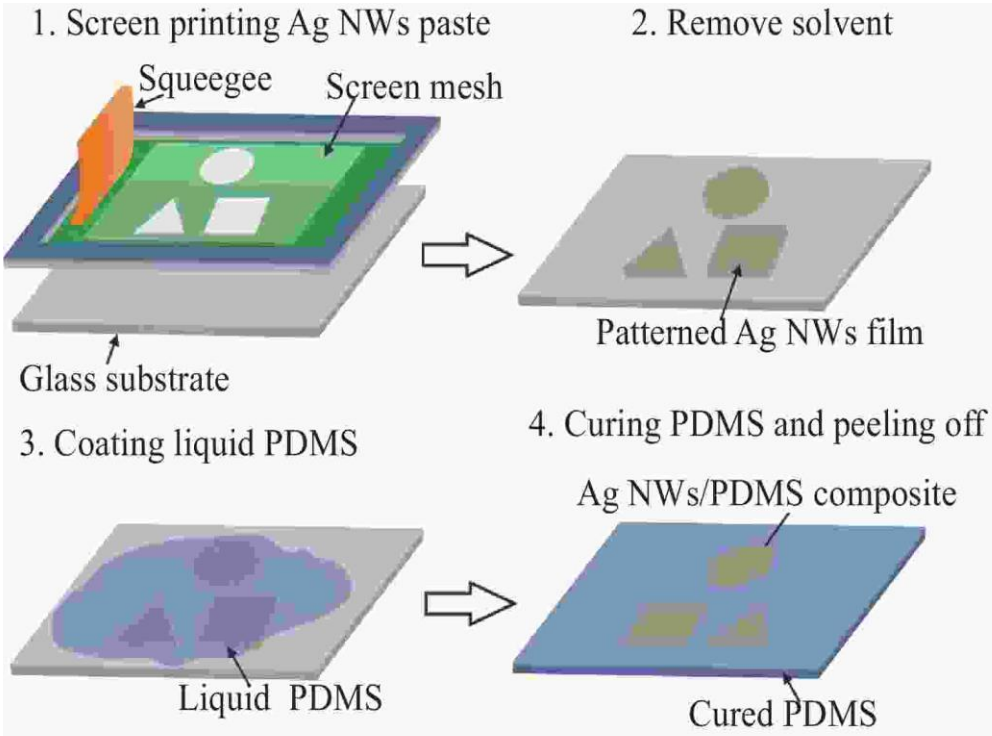

Figure 2.6 Schematic illustration of the fabrication process of patterned stretchable circuit (Yuan *et al.*, 2018)

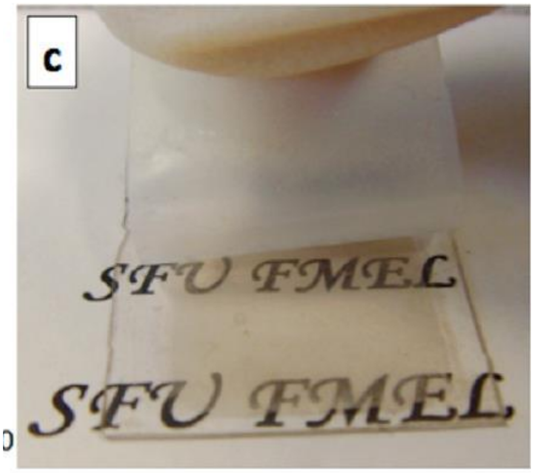

Figure 2.7 The enhanced adhesion of the AgNWs to the dopamine- modified PDMS surface was monitored using the taping test (Akter and Kim, 2012).

2.5 Conductive Ink

The conductive ink is a mixture of metallic particles dispersed within a solvent. The non-Newtonian rheology of this mixture allows the ink to be screen printable which is preferable in the fabrication of stretchable electronics in varies of designs (Mohammed and Pecht, 2016). Generally, there are three notable metal (Au, Ag and Cu) particles used in the fabrication of the conductive ink. These conductive materials would be the first element of the conductive ink which directly determine the functionality of the conductive ink in term of electrical conductivity.

The second element of the conductive ink would be the polymer binder. This polymer binder plays an important role in attaching the metallic particles to the substrate during and after the sintering process. The final element of the conductive ink would be the volatile solvent and non-volatile organic polymer which generate the rheological properties in the ease of screen printing.

In the fabrication of stretchable electronics, conductive ink is important to have the capability to be stretched by at least 20% of its original length. Besides, after multiple cycles of stretching, the conductive ink is required to remain its electrical and mechanical integrity.

2.6 Mechanism of Stretchable Electronics

In the various applications, stretchable electronics respond to the different level of applied strain with different mechanism. Unlike the traditional strain gauges, mechanism such as disconnection between piezoresistive effect, percolation mechanism, tunneling effect, disconnective mechanism and crack propagation have been utilized to study and develop behaviour and functionality of the stretchable electronic (Amjadi *et al.*, 2016).

2.6.1 Piezoresistive Effect

The change in the electrical resistivity of the materials which is caused by the structural deformation is known as the piezoresistivity. The variation of the electrical resistivity is induced due to the dependence on mobility and density of the charge carrier. Therefore, when there is stress and strain applied on the piezoresistor, the stress and strain may modify the width of the band gap and the mobility of the electrons and holes which are the charge carriers.

Generally, the piezoresistors work by the mechanism of piezoresistive effect which is popular in the development of stretchable pressure and strain sensors (Trung and Lee, 2017). Traditionally, the piezoresistors is made by metals, metal alloys or semiconducting materials such as silicon and germanium which is able to present a higher resistivity variation that respect to the change of resistance induced by the geometrical changes (Stassi *et al.*, 2014). However, there is a major drawback from these semiconducting and metallic piezoresistors which are the fragility and rigidity. In addition, these semiconducting and metallic piezoresistors are costly to be fabricated and very sensitive to temperature. This issue lead to the development of development of stretchable conductor materials with high piezoresistivity which is commonly fabricated in the form of composite of conductive filler with the elastic polymer matrix (Trung and Lee, 2017).

In addition, the nanoscale material such as AgNWs, AgNPs, CuNWs and CNTs are used to fabricated stretchable electronic with ultrahigh piezoresistivity due to the chirality and change of barrier height (Amjadi *et al.*, 2016). However, there is a concern about the contribution of the piezoresistivity of these nanomaterials on the stretchable electronics may be low. It is because there is a problem about the compatibility of the elastic polymer with the nanomaterials. Poor compatibility of the piezoresistivity nanomaterials and elastic polymer may lead to poor interfacial adhesion and yield negligible deformations of the nanomaterials upon stretching.

2.6.2 Percolation Mechanism

The variation conductivity of nanocomposite is the result of the variation of conductive filler concentration. It is because the conductivity is governed by the percolation mechanism, also as known as percolation theory. The percolation theory can be expressed in:

$$\sigma \approx \sigma_o \ (p - p_c)^t, p > p_c \qquad \text{(Equation 2.1)}$$

Where σ is the bulk conductivity of the composite, σ_o is the conductivity of the filler, p is the weight percentage of the filler and is the critical exponent. The critical percentage p_c of filler is defined as the percolation threshold (Dang *et al.*, 2017). A continuous electrical pathway is built when the concentration of the conductive nanofillers has exceed the percolation threshold and achieve the critical fraction which is illustrated on Figure 2.8.