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## The study of non-permanent tattoo ink using nano silver compounds and untact 3D printing technology

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THE STUDY OF NON-PERMANENT TATTOO INK  
USING NANO SILVER COMPOUNDS AND  
UNTACT 3D PRINTING TECHNOLOGY

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Master's Program in Metallurgical and Materials Engineering

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2020

THE STUDY OF NON-PERMANENT TATTOO INK  
USING NANO SILVER COMPOUNDS AND  
UNTACT 3D PRINTING TECHNOLOGY

by

JISU LEE, BS

THESIS

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DEPARTMENT OF METALLURGICAL, MATERIALS and BIOMEDICAL ENGINEERING

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## **Abstract**

Due to a recent epidemic of COVID 19, the possibility of 3D tattoo printing technology using the Internet of Things (IoT) control system developed in a non-face-to-face contact process was identified. A non-surgical tattoo process and hygiene safety differentiated with silver tattoo ink was used to solve the health problems arising from the surgical tattoo process and to identify 3D tattoo printing techniques through a non-personal contact concept. During the tattoo processing, the image selected by the customer is sent to a designated printer via an IoT control system in a separate location. A piston-type extruder (PTE) printing technique was used with a 100-micrometer nozzle tip. The tattoo ink was printed on a hydrophobic or hydrophilic substrate, and then tightly contacted to a stratum corneum composed mainly of keratin and lipids. Based on the thickness of the stratum corneum diffused from the surface according to the replacement cycle of 2 weeks, it was confirmed by the tattoo ink concentration and contact time that the printed tattoo disappeared within 3 days to 2 weeks.

The silver tattoo ink disappears entirely in two weeks after it has been elaborately transferred to the stratum corneum of the human skin or nails in a web-based, non-face-to-face contact printing process. Repeated testing has proven that painless attachment to prints and sterile surfaces is an active process to control the quality of non-permanent tattoos. IoT enabled tattoo printing technology developed from the non-face-to-face contact concept includes the following steps: 1) Preparation of silver tattoo ink; 2) Accurate dispensing control of 3D printed materials; 3) Unique and fast design; 4) Convergence of web-based control to prevent the transmission of infectious diseases and protect personal information.



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## **Chapter 1: Tattoo Technology In Dermatology**

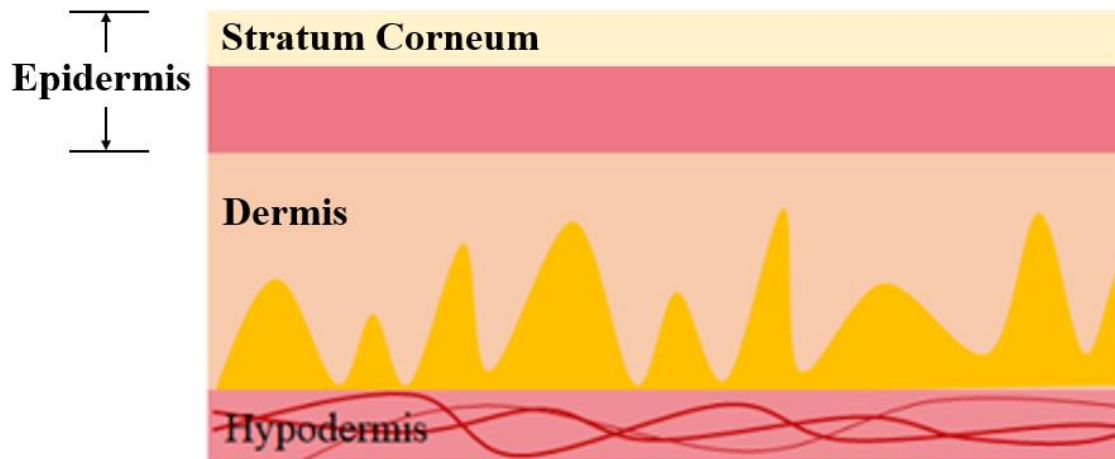
In modern society, the desire for expression spreads rapidly, regardless of age and gender. According to the results of the online survey of The Harris Poll in 2015, 29% of Americans have at least one tattoo. Compared to the same survey in 2011, roughly 21% of people got tattoos in America. It can be seen that the number of people who gets tattoos increased sharply [1]. The surgical tattoo procedures should be done by medical practitioners from the three significant different areas of beauty-art, including hair, makeup, and nail art. Tattoo art has been recognized as an elaborate medical practice. The role of tattooists was shifted by 3D printing tattoo technology from the artist field to the scientist domain with the use of 3D printer. In this paper, it is clearly explained in detail with the development of economical equipment and the printing techniques.

### **1.1 Structure of Human Skin**

Skin is the largest organ and covers all surfaces of body. The skin of adults weighs around 8 pounds and has an area about 22 square feet [2]. It protects body and helps to control body temperature, which is made up of three layers.

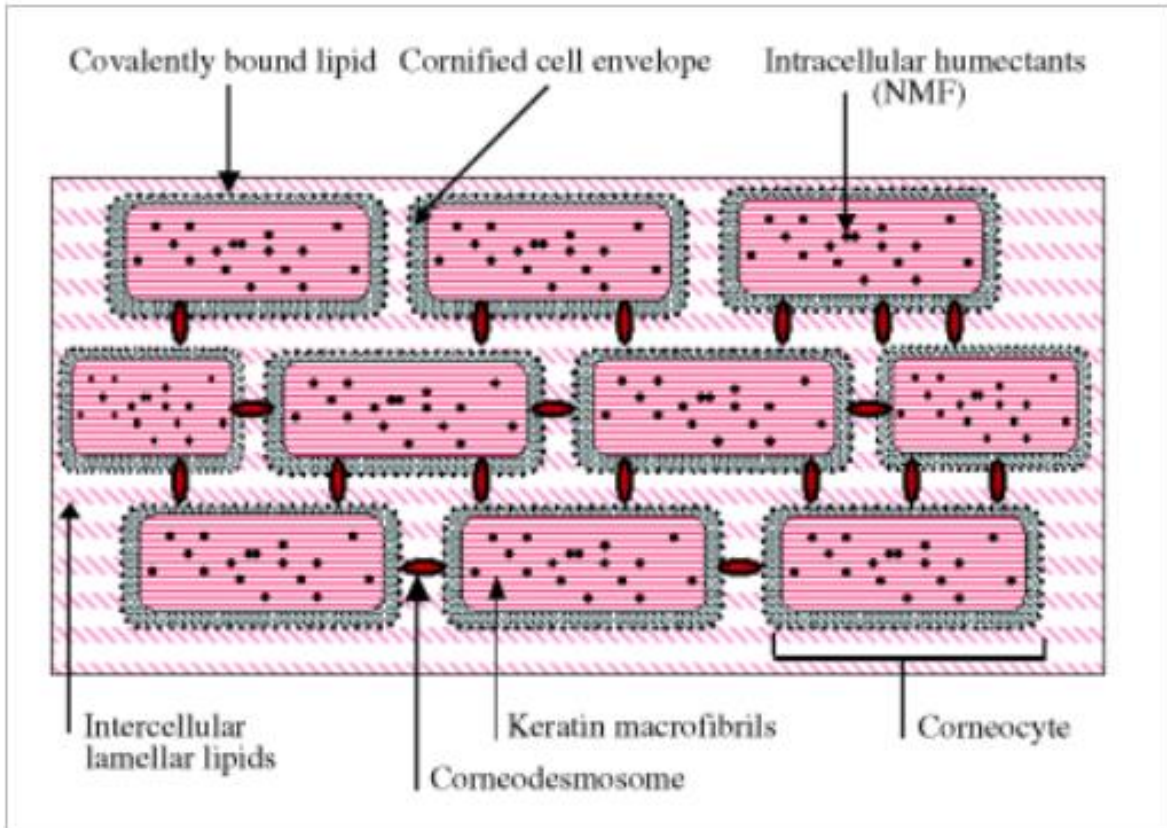
#### **1.1.1 The Structure of Human Skin Layers**

Human skin is made up of three layers consisting of epidermis, dermis, and subcutaneous tissue or hypodermis, as described in Figure 1.1 [3]. Epidermis is the outer layer in skin, which defends the living tissues as a primary barrier against outer dangers including infection, injury, chemical and mechanical stresses, and dehydration. The epidermis has five layers: stratum basale, stratum spinosum, stratum granulosum, stratum lucidum, and stratum corneum.



**Figure 1.1 The layers of the skin: Epidermis (with Stratum Corneum), Dermis, and Hypodermis.**

The important layer is the stratum corneum that is the outer layer of epidermis [4]. It is made up of keratin and lipids. On the Figure 1.2, the structure of stratum corneum is similar to a wall with brick and mortar in which non-continuous matrix of keratin are embedded in continuous matrix of lipids [5]. Stratum corneum layer and nail have a role of body temperature regulation, protection from hazards, prevention water loss or absorption, and cell renewal following its cycle [6-8]. Furthermore, the color of human skin is determined by melanin pigment which are produced from melanocytes cell. The melanocytes are developing in the epidermis [9]. Dermis is under the epidermis, which includes tough connective tissue, cells, glands, and blood vessels. Subcutaneous tissue is called hypodermis, which is located under the dermis. It help attach the skin to blood vessels, nerves, bones and muscle, which consists of fat and connective tissue [10].

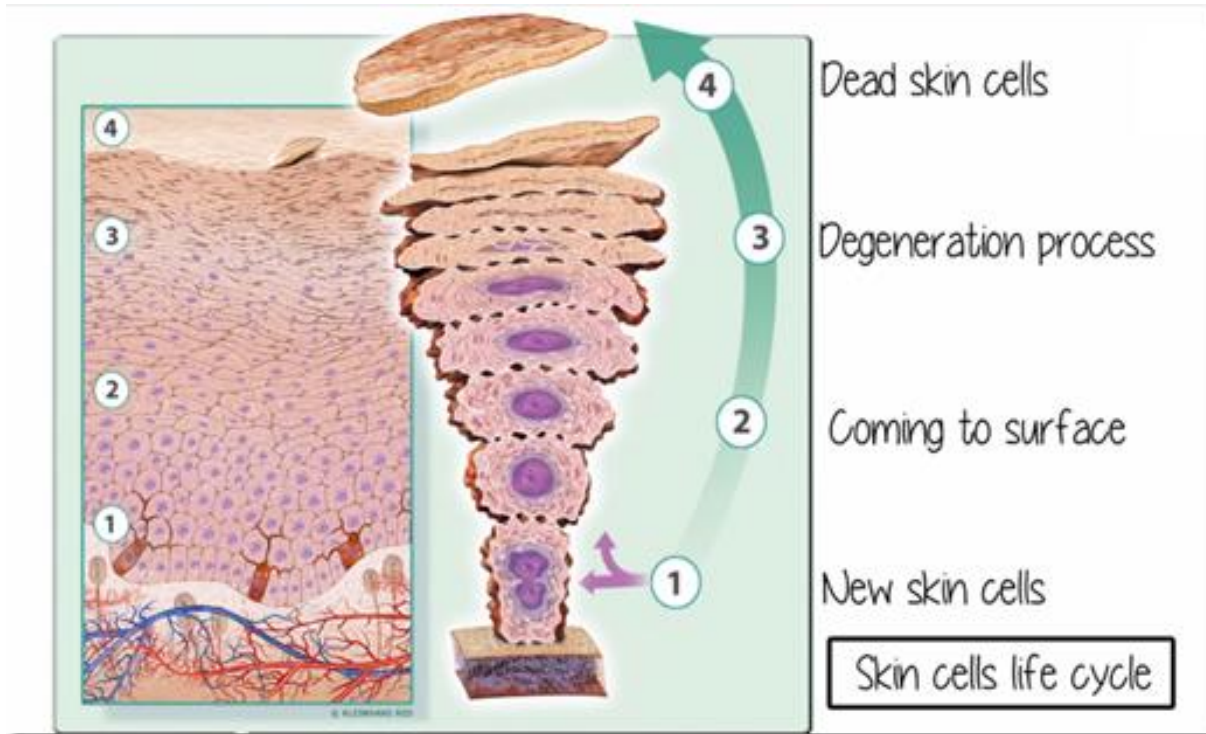


**Figure 1.2 The structure of stratum corneum with keratin and lipids. [5]**

### **1.1.2 The Human Skin Cells Life Cycle**

A new skin cell is created at the deepest layer of the epidermis and grown up to the surface of the skin, and the dead skin cell has peeled at the stratum corneum. The cycle is repeated, which is called a skin cells life cycle (described in Illustration 1.1). The average skin cell cycle for humans in the epidermis is 5 to 6 weeks, but it varies in individuals, skin conditions, age, and hormones. The skin cycles of the age of 19 to 21 is 2 to 3 weeks and that of the age of 50's and 60's is 8 to 13 weeks. The skin cycle gets slower as people are older [11]. However, the cell cycle in the stratum corneum is quite different from the skin cycle in the epidermis. It is a 2 week cycle, considering the cell cycle within the stratum corneum. A keratinocyte enters from the deeper layers of epidermis to the stratum corneum and changes into corneocyte and shed for 2 weeks [4].





**Illustration 1.1 Skin cells life cycle for human. [11]**

A needle for permanent tattoo penetrates through the outer layer of human skin with puncture skin at a frequency of 50 to 3000 times per minute. When the tattoo needle penetrates the skin, it causes scars or wound and can be infected from any viruses. And its chemical ink particles flow into the epidermis and dermis layers which contains nerves and blood vessels [12]. Different from the permanent tattoos, nails and stratum corneum layer are areas where tattoos inks can be applied without alive cell activities such as a neuron, DNA, and RNA.

## **1.2 Conventional Tattoo Technology in Human Skins and Cells**

Conventional tattoo technology is related to a permanent tattoo or a temporary black henna tattoo. People usually like the permanent tattoo and the black henna tattoo, but there are some limitations such as hygiene, chemical ink, and further removal procedures. And also, there is no distinct regulation that surgical tattoo procedures should be done by medical practitioners from the

three significant different areas of beauty-art, including hair, makeup, and nail art. Tattoo art has been recognized as a delicate medical practice due to the invasive process of the permanent tattoo.

### **1.2.1 Permanent Tattoo**

For the permanent tattoo, the tattoo injects chemical ink into the epidermis and endothelial area which line the interior surface of blood vessels and lymphatic vessels in human skin. That means the needle for permanent tattoo breaks the skin barrier treating living tissue [13]. Based on hygiene problems, only clinic professionals are allowed to do the tattoo and removing clinics [14]. Both tattooists and the customer who is treated by non-certificated tattooists could be perceived as potential criminals from perspective of a health risks. Scientifically sufficient data and researches report hygienic problems. 2 % of the US population is infected by Hepatitis C, and 33% of the infected have done tattoos [15]. Researchers reported that the tattoo ink dissolved in the blood and circulated in the vessel, and heavy submicron-particles are deposited in lymphoid organs [16]. Moreover, tattoo procedures are invasive to human skins. It occurs infections, allergies, and disorders in the human immune system because of the surgical procedure being conducted under non-hygienic environments [17-22].

Fortunately, the tattooists have obligations to be trained about sepsis and hygiene in France since 2009. Sepsis is the response of body to an infection [23]. Due to unsanitary environments and medical issues of tattoos, the government of South Korea has restricted any tattooing by non-certificate tattooists. However, only a few doctors have tattoo skills because there are artistic limitations and is a lack of educational opportunity for necessary tattoo skills. As the regulations on tattoos become stricter and harder, the number of home tattooists or unlicensed tattooists is increased in secret. An unsafe and ironic situation was occurred due to regulations or obligations

for safety tattoos. Therefore, a new paradigm is suggested in this paper to develop a novel tattoo process with the help of scientific knowledge.

14% of people who got tattoo experienced tattoo-related regret in the United States. The main reasons for regretting those could be health risks and permanent scars [24]. At first, the health risks including infection and allergic reactions make people who got tattoo regret. Those health risks can be resulted from two possible reasons: non-hygienic environment as mentioned before and the allergic reactions to chemical mixtures in tattoo inks [25]. Tattoo inks consist of two components: the carrier and the pigment. The common carriers are composed of water, glycerin, isopropyl alcohol, and witch hazel. The carrier works as a suspension to maintain the pigment well mixed and protects from pathogens [26]. The tattoo inks are not safe, but manufacture companies do not have any requirements or obligations to inform about the ingredients of the tattoo inks. The tattoo inks could be included harmful ingredients such as dirt, rust, metal salts, plastics, or soot [27]. In addition, the pigments of the tattoo inks for colors are made of heavy metals: nickel for black color and lead for yellow, green, and white colors. The details of the heavy metals and their colors are summarized in Table 1.1 [28]. A 43-year-old person got tattoo and formed squamous cell carcinoma (SCC) at the tattoo region after three months [29] (Figure 1.3).

**Table 1.1 Heavy metals used for Tattoo ink colors [28]**

Heavy Metals	Colors
Mercury	Red
Lead	Yellow, Green, White
Cadmium	Red, Orange, Yellow
Nickel	Black
Zinc	Yellow, White
Chromium	Green
Cobalt	Blue
Aluminum	Green, Violet
Titanium	White
Copper	Blue, Green
Iron	Brown, Red, Black
Barium	White
Metal Oxides (Ferrocyanide and Ferricyanide)	Yellow, Red, Green, Blue
Organic chemicals with Azo-chemicals (N)	Orange, Brown, Yellow, Green, Violet
Naptha-derived chemicals	Red
Carbon (Soot or Ash)	Black



**Figure 1.3 Squamous cell carcinoma (SCC) formation at tattoo region. [29]**

Secondly, numerous people regret getting the permanent tattoos, due to permanent scars. The more impulsive, the more tattoo regret people experienced. After getting the permanent tattoo

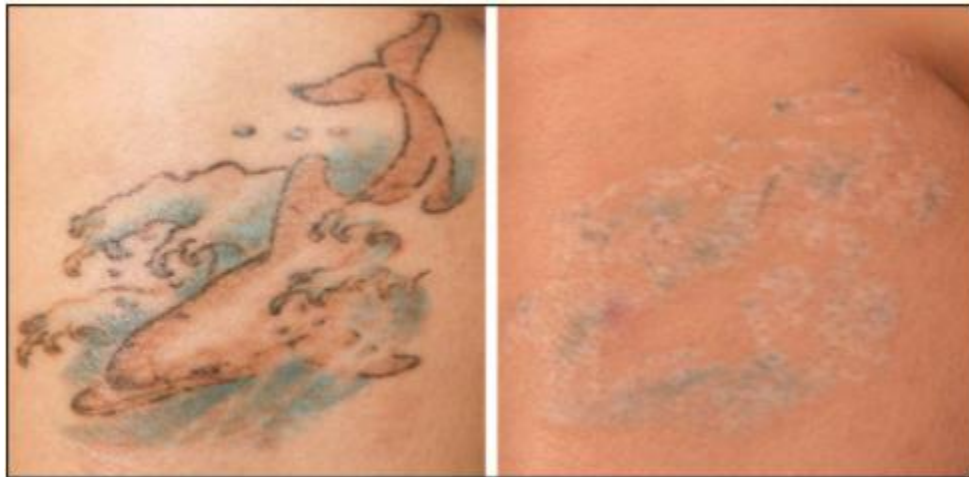
onto skin, it is difficult to remove or change the shape of tattoos. This is because tattoo needle penetrates through the outer layer of human skin with a wound and deliver its chemical ink into the epidermis and dermis layers which contains nerves and blood vessels. Removal of tattoo is available with multiple laser treatments, but scars are left due to irregular depths of the pigmented skin layers [30-32]. During the laser tattoo removal procedure, the tattoo ink particles are heated by the laser light under the skin surface like the epidermis and dermis layer. And the laser light makes the tattoo ink particles split into the smaller ink fragments [33].

The smaller ink particles are destructed by body immune system such as macrophages and lymphocytes. They are managing the immune system which protects the human body from invaders. Macrophages eat the invaders as an innate response and transport to lymphocytes to destroy them as adaptive response. Additionally, B-lymphocytes detects the invaders while T-lymphocytes destroy the invaders using lymphatic fluid. The concentration of the smaller pigment particles decreases in skin, and the color can be cleared from the human skin by body immune systems. The original ink particle is too large to be eaten by the macrophages. That is why the permanent tattoo inks keep staying in dermis layer [34].

According to the result of the online survey, only 38% of people complete the laser tattoo removal treatments [35]. The removal of tattoo process is more painful, time-consuming, and more costly than acquiring the tattoo. The cost of the laser tattoo removal depends on several factors such as the size, color and the amount of the years of the tattoo. The average cost varies from \$65 to \$540 per laser tattoo removal treatment session. Since the laser tattoo removal treatment does not end with a single procedure, the total amount could increase exponentially [36]. Moreover, the laser tattoo removal process can cause unwanted effects including scars, dyspigmentation, ink darkening, and poor skin conditions like nonrecovery of skin color [37-39] (Figure 1.4-1.6).



**Figure 1.4 A black colored tattoo treated with multiple treatment of laser. A) Before and B) After 2 sessions of the laser tattoo removal treatments. [37]**



**Figure 1.5 A multicolored tattoo treated with multiple treatment of laser. A) Before and B) After the laser tattoo removal treatments. [37]**



**Figure 1.6 Achromic laser tattoo scars: A) After 18 laser tattoo removal treatment sessions and B) After 23 laser tattoo removal treatment sessions. [37]**

### **1.2.2 Temporary Black Henna Tattoo**

As an alternative to a permanent tattoo, temporary henna tattoo has been developed. However, some people get sick from allergies after getting henna. Actually, a contact dermatitis with allergic reactions is not caused by natural henna, but by coloring additives in black henna tattoo mixtures [25] (Figure 1.7-1.9). The additives which increase the allergic reactions are para-phenylenediamine (PPD) and heavy metals. There are some cases that have been reported. One of the reported cases is about PPD. PPD is used as a sensitizer, which accelerates the dyeing process. It makes the color darker and also occurs an allergic contact dermatitis, which is considered as an allergen [40-43].

A 37-year-old woman dyed her hair with the black henna and became in her scalp. An allergic test was performed on her arms with the black henna tattoo and 1.0% of PPD in petrolatum



to make sure the allergic reactions. Those developed the contact allergic reaction and pruritic symptoms [44] (Figure 1.10). This test was based on 1.0% of PPD, but the commercial black henna tattoo samples purchased from tattoo shops contain PPD at levels from 3.37% and up to 51.6% [45].

The other reason that causes the contact allergic dermatitis is due to heavy metals in black henna. The heavy metal levels in the black henna tattoo ink were found to be 0.44–3.11 ppm for Co, 1.13–2.20 ppm for Ni, 1.59–17.7 for Pb, and 35.0–76.9 ppm for Cr. The details of the levels of the heavy metals are summarized in Table 1.2. The heavy metal contaminants in black henna tattoos increase the risk of allergic reactions and contact dermatitis [45]. Strict regulations for PPD and the heavy metals in the tattoo inks are required to regulate their concentration.



**Figure 1.7 Allergic reaction to black henna on face, back hand and arm. [25]**

**Figure 1.8 Keloidal reaction to black henna on face. [25]**

**Figure 1.9 Keloidal reaction to black henna on arm. [25]**





**Figure 1.10** Test results to 1.0% of PPD in petrolatum and black henna tattoo mix on arm at 24 hours. [74]

**Table 1.2** The levels of the heavy metals in temporary black henna tattoos. [45]

Sample	Heavy metals (ppm)			
	Cobalt	Nickel	Lead	Chromium
1	$1.89 \pm 0.58$	$1.40 \pm 0.35$	$9.16 \pm 2.07$	$52.29 \pm 5.81$
2	$3.11 \pm 1.35$	$2.20 \pm 0.82$	$17.7 \pm 10.24$	$76.9 \pm 21.83$
3	$1.04 \pm 0.001$	$1.13 \pm 0.17$	$7.08 \pm 0.76$	$55.25 \pm 2.13$
4	$0.74 \pm 0.61$	$2.10 \pm 1.17$	$6.69 \pm 0.8$	$66.8 \pm 19.04$
5	$0.44 \pm 0.35$	$1.70 \pm 0.91$	$1.59 \pm 0.49$	$35.0 \pm 47.42$

### 1.3 Silver Ink Tattoo Technology

As mentioned at the 1.2 Section, the ingredients of the commercial tattoo inks for the permanent tattoo and non-permanent black henna tattoo are harmful to human. On the contrary, 3D printable tattoo uses silver compounds as a tattoo ink in this thesis. Based on real applications

of the silver compounds in biological parts, it is known that the silver compounds are stable and biocompatible materials.

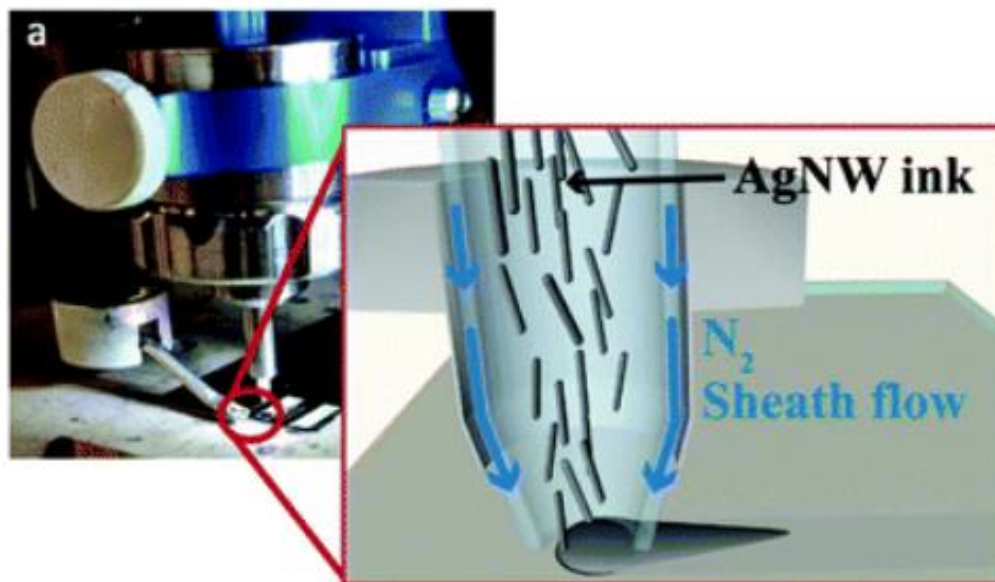
In this paper, refined silver tattoo inks are dispersed in the depth 10-100 $\mu$ m of the stratum corneum layer. Consideration of tattoo material applying 3D printing method is necessary for the solution of health problems, social problems between medical professionals and tattooists, and artistic limitations. The non-permanent tattoo could be recognized as a cosmetic field, not as a medical field. Along with the fundamental problems and recent hygienic issues of the tattoo process, 3D printing technology opens up the possibility of completely changing the global tattoo market in the future.

### **1.3.1 Stability of silver compounds**

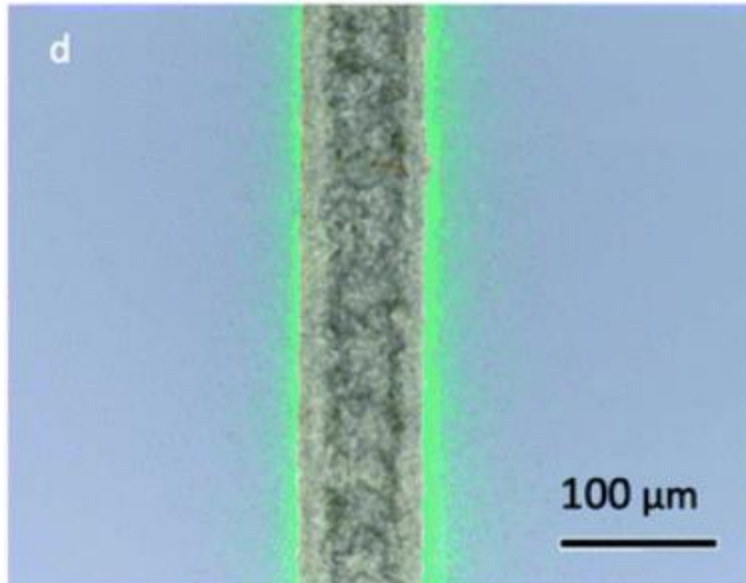
Nano-silver powder has been proved to be harmless to the human body as it is utilized as biological applications such as an electronic ink, a disinfectant and a treatment. One of the biological utilities of the nano-silver components is silver nanowire inks. The silver nanowire inks are used to epidermal electronic ink tattoos, which are biological electronic devices containing technology-equipped biosensors onto biological tissue such as the human skin. It has a general tattoo shape, which acts as a wearable watch and can be controlled with a smartphone. Figure 1.11 indicates that the deposition method for the silver nanowire inks was aerosol jet printing at room temperature. And the actual images of the electronic tattoo ink are presented in Figure 1.12 and Figure 1.13 The water-based inks consisting of high aspect ratio silver nanowires have high biocompatibility without harmful post-processing [46].

Another biological utility of the nano-silver components is a disinfection. A nano-silver compounds are used to filter contaminants in water as a disinfection for drinking water [47]. And silver nitrate compounds are used for clinical purposes such as treatment of infections in burns or

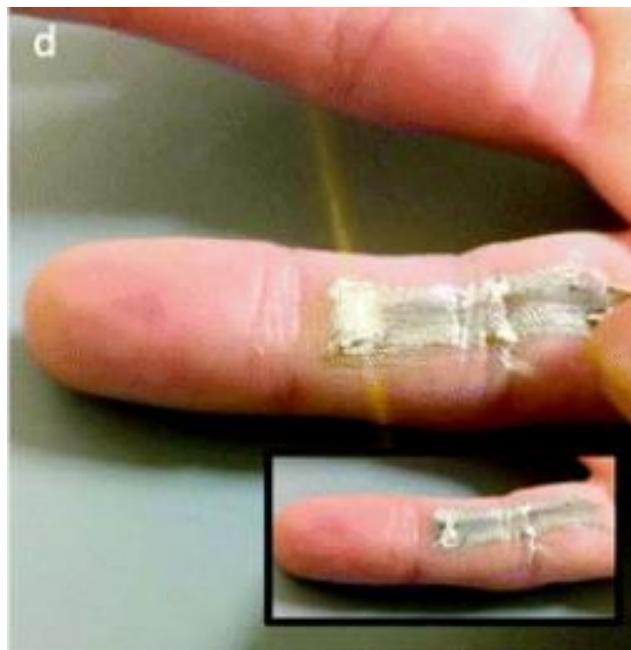
wound, due to inhibitory of bacterial growth activity and its great stabilization and biocompatibility [48-49]. In 2015, a researcher found that plant extract derived silver nanoparticles have remarkable antioxidant and anticancer properties. The silver nano compounds could be used to treat serious diseases such as cancer [50-52]. Ink using silver components needs further research to make harmless for allergic users. In this way, silver compounds have been used in humans for therapeutic purposes, disinfection and biological electronic devices with great biocompatibility.



**Figure 1.11 Aerosol jet printer head and the printer nozzle with silver nanowire ink. [46]**



**Figure 1.12** Optical image of a printed silver nanowire ink trace on glass. [46]

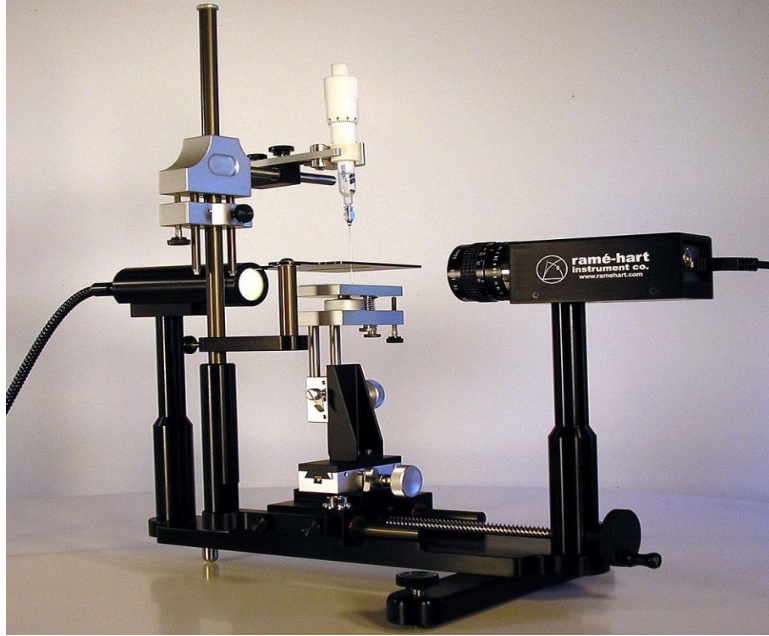


**Figure 1.13** The electronic tattoo was printed on a finger and deliver power to an LED. [46]

### 1.3.2 Silver Tattoo Ink and Contact Angles

Contact angle where liquid-vapor interface meets a solid surface can define wettability of solid surface with different liquids. The contact angles are measured by Sessile drop method with contact angle goniometer in Figure 1.14. The contact angle of hydrophilic materials is smaller

than 90°. And the contact angle of hydrophobic materials is larger than 90° [53]. A material which has a less contact angle can easily make wet the surface. It means that the hydrophilic materials can be absorbed easily, and the hydrophobic materials are not.



**Figure 1.14 Sessile drop method (Contact angle goniometer, Ramé-hart instrument co.)**

In the study of the 3D tattoo printing, the hydrophobic material is the collagen microneedle, and the hydrophilic material is silver nitrate tattoo ink. Based on the wettability concepts, the tattoo ink cannot disperse into the epidermis and dermis layers because of the collagen microneedle which is located in epidermis layer. The hydrophobic property of the collagen microneedle controls the diffusion rates of the silver ink into the skin. The ink only spreads in the stratum corneum and maintains during the 2 week-cell cycle.

### **1.3.3 Ink Diffusion Rates (Fick's 2<sup>nd</sup> Law)**

Fick's laws of diffusion (Fick's 2<sup>nd</sup> law) were derived by physiologist Adolf Fick in 1855. The law describes diffusion rates with the mathematical equation  $\frac{\partial C}{\partial t} = -D \frac{\partial^2 C}{\partial x^2}$  (C: Concentration, t: contact time, D: diffusion coefficient, and x: the position or the distance of

location in skins). It predicts how diffusion causes the concentration to change in terms of time. The solute molecules diffuse to fill the space when removing the barrier between the spaces [54]. In the thesis, the solute molecules can be the silver nitrate inks, the barrier can be the skin barrier, and the reservoirs can be the stratum corneum. The simulation of diffusion of silver ink in skins followed a Fick's 2nd law, which describes the diffusion rates of the silver tattoo ink depends on the ink concentration and the distance in skins.

Based on the Fick's 2nd law, the penetration rates of keratin in the stratum corneum control the silver tattoo ink concentration and its diffusion rate, and the tattoo ink is not absorbed into the dermis. The silver ink tattoo experiment was designed to release the ink along with the 2 week-cycle of the stratum corneum. The amount of silver ink is decreasing as the skin gets more rooted within the stratum corneum. The diffusion rates depend on the exact penetrated distance of the ink in skins.

The varying factors are the different concentrations of the silver ink after the contact time with varying diffusivity  $D$  values. And the moisture and oil contents of different locations of skin, thicknesses, and temperature, different printing conditions can be factors that changes the dispersing speed rates and tattoo duration period. The projected non-permanent silver ink tattoo shape, the duration period and the removal speed of the tattoo on the stratum corneum depends on the dispersing speed rates.

#### **1.3.4 Fabrication of Microneedle Patch**

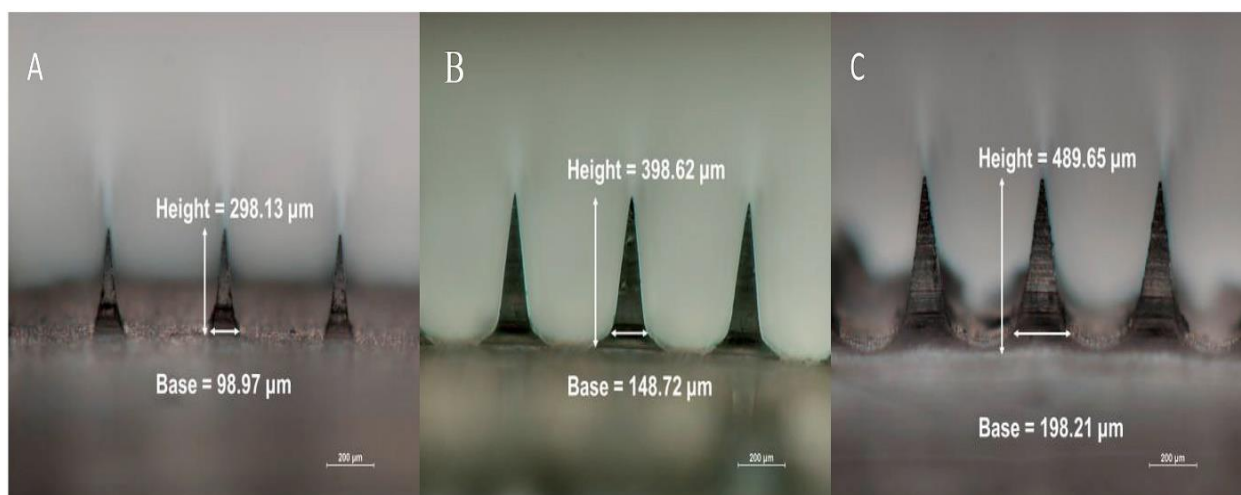
A microneedle patch is composed of micron-sized needles, which is sterilized and penetrates the skin using the drug delivery systems for treatment as pharmaceutical uses and cosmetic purposes. The microneedles were made of collagen-based biopolymers which have biodegradability, biocompatibility and non-toxic properties [55]. Collagen is known as

composites for repairing bones, teeth, skin and tissues, which are the major solid components of the human bone and teeth [56].

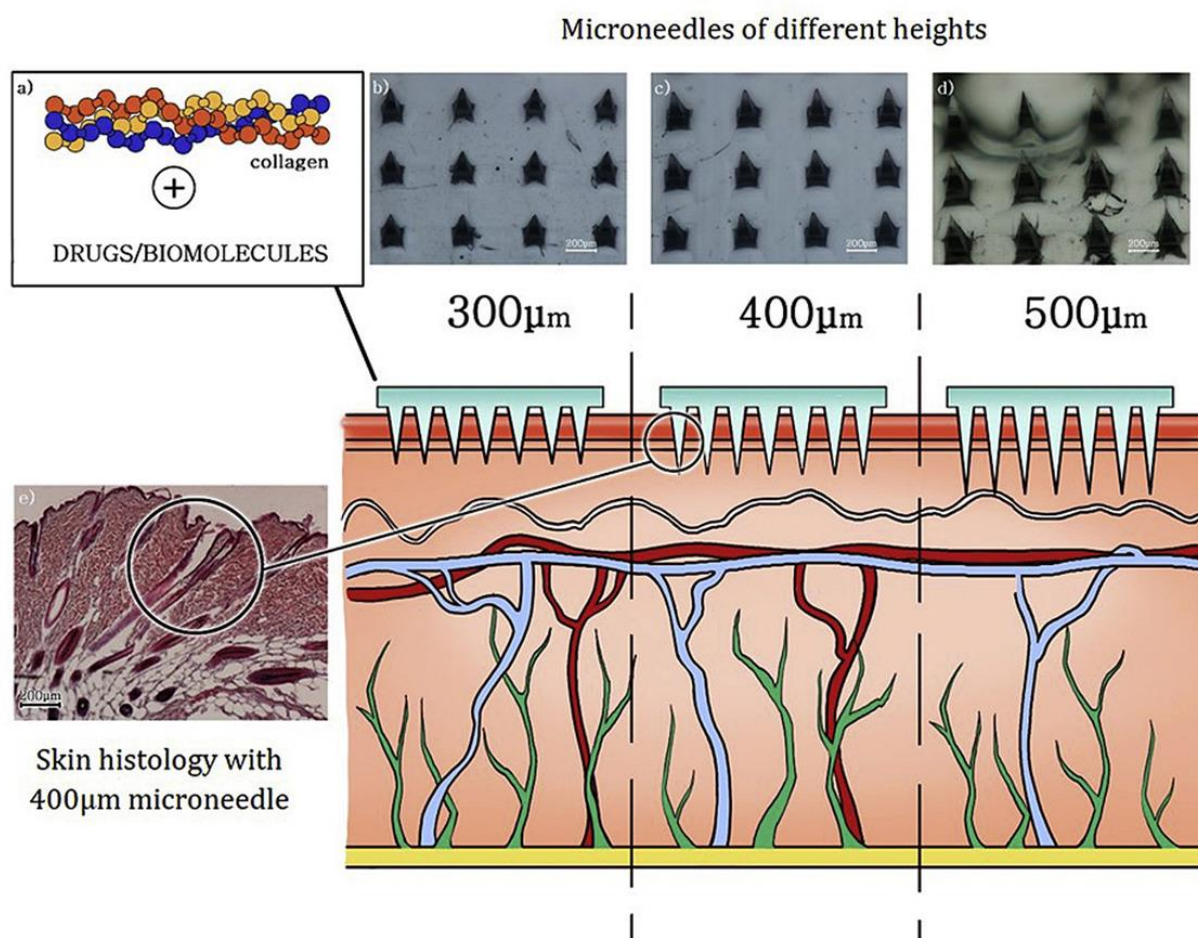
An experiment to penetration of the collagen microneedles in mice skin was performed in the University of Texas at El Paso (UTEP). The uniform collagen microneedles are synthesized with polyvinyl pyrrolidone (PVP) by micro-molding technique that is a simple, economic technology. PVP strengthens the collagen microneedles to sustain the three-dimensional forms. Hydrolyzed collagen and PVP are filled into Polydimethylsiloxane (PDMS) microneedle molds to manufacture the collagen-based microneedle patch. The production process for the microneedle patch was placed inside a vacuum oven below atmospheric pressure for 12, 18, and 24 hours. The microneedle patches will be removed from the PDMS molds when the patches were dried [57].

The Figure 1.15 shows the produced collagen-PVP microneedle patches using micro-molding technique with various heights: 300, 400 and 500  $\mu\text{m}$ , and the bases: 100, 150 and 200  $\mu\text{m}$ . The Illustration 1.2 indicates the diagram of epidermal layer in skin with penetrating of the collagen-based microneedle patches which have different depths and bases. The heights of the microneedles determine the penetration location of the silver ink. The microneedle patch can penetrate the stratum corneum and the epidermal layer, but not penetrate into the dermal layer [57]. The collagen microneedles can deliver and dissolve the drugs or the nutrients exactly to the expected or targeted points in skin with varying the depths of the needles [58].





**Figure 1.15** Collagen-PVP Microneedle Patches with various bases and heights: A) 100  $\mu\text{m}$  x 300  $\mu\text{m}$ , B) 150  $\mu\text{m}$  x 400  $\mu\text{m}$ , and C) 200  $\mu\text{m}$  x 500  $\mu\text{m}$ . [57]



**Illustration 1.2** The diagram of epidermal layer in skin with penetrating of the collagen-based microneedle patches which have different depths and bases. [57]



### 1.3.5 Structure of Skin Layers and Microneedle Silver Ink Patch Tattoo

Actually, it is difficult to maintain the tattoo shape of the silver nitrate ink onto the skin or substrate. This is because the silver tattoo ink is in a liquid state if the ink is sprayed itself directly onto the skin. Even if the tattoo ink is printed along the tattoo design from the 3D printer, the liquid ink may not keep the shape and be absorbed into the skin. The spilled ink can ruin the expected tattoo design, and even last for two weeks with ruined tattoo design. To avoid the bad situation, it needs something to maintain the shape of the tattoo ink, to control the dispersing rates of the silver ink and to reach exact depth along the tattoo design. The answer key for this situation is attaching a microneedle silver ink patch tattoo.

The attaching microneedle silver ink patch tattoo can be a drug delivery paradigm. The mechanism of the silver tattoo ink is similar to the mechanism of the drug delivery system which has to control the diffusion of the dissolved drug molecules through the controlling element for diffusion rate. The drug is delivered to targeted receptor like a specific part of body, while the silver ink is diffused to targeted design [59]. And they have the same purpose for slow release of the drug or the silver ink to the body at the constant diffusion rates.

The tip of the collagen needle is discolored by silver nitrate ink. The micro collagen needle patch was utilized to make the similar conditions such as contents of moisture and oil for a different type of skins or the skin of different individual people. The silver ink was dispersed on the stratum corneum and epidermis layers of the skin layers with microneedle. The hydrophobic collagen microneedle dissolved in epidermis layer, which is located in the depth 300-600 $\mu$ m from the skin surface. It does not contact dermis layer, because the depth of the microneedle is less than 5 $\mu$ m. The hydrophobic microneedle protects the hydrophilic silver ink to penetrate into the dermis layer. And hydrophilic silver nitrate ink spreads into the stratum

corneum layer, where embedded non-continuous matrix of keratin in continuous matrix of lipids. The refined silver tattoo inks are dispersed into the stratum corneum layer which is located in the depth 10-100µm from the skin surface. The ink is be applied to the layer without alive cell activities.

Due to the hydrophobic properties of collagen microneedle patch, the tattoo ink does not diffuse into the epidermis and dermis layers. The hydrophilic silver ink spreads only in the stratum corneum, and its dispersing speed is controlled by the collagen microneedle. The hydrophobic property can control to slower the silver ink penetration rates into the skin and allow the ink to stay in the layer during the cell cycle. After diffusion of the silver tattoo ink, the printed shape maintains in the stratum corneum during the 2 weeks of the cell cycle. Using the disposable microneedle silver ink patch provides the hygiene condition and economic tattoo process.

## **Chapter 2: 3D Biomedical Printing Technology**

In response to the recent global pandemic of coronavirus 2019 (COVID-19), a public health activity guidance has forced people to restrict the face-to-face contact or interaction among individuals. Social distancing is vital to alleviate the spread of the coronavirus and is a long-established public health tool for the infectious agents [60]. The instructions are that limitations on gatherings and the operation of businesses, individuals maintain a distance from one another in public, and suggestion to stay at home [61]. The COVID-19 pandemic has had tremendous negative impacts on the public health and global economies [62]. The instructions are essentially applicable to tattoos and beauty-art fields [63]. To address the ongoing COVID-19 pandemic, the contactless systems have emerged and are being developed.

3D printing is one of the untact system which has the same meaning to contactless system that can be controlled from outside. For the permanent tattoo or temporary black henna tattoo, the tattooist and the customer are in close contact with each other with sharing the same space for more than 30 minutes or an hour because the tattoos are handmade. Getting tattoo is a very dangerous procedure where the social distancing is impossible, requiring a distance more than 6 fts apart. Tattooists repainted design by hand, which took a long time and not reproduceable. However, 3D printed tattoo is different. The possibility of 3D tattoo printing technology using the Internet of Things (IoT) control system developed in a non-face-to-face contact process was identified [64]. In a separate place, users can remotely and precisely draw designs through IoT control technology or send designs from photos, existing files, or web-based programs. The technology has been attempted to print the selected files onto the substrate.

### **2.1 Conventional 3D Printing Technology**

When digital files are transmitted from an inkjet printer in the daily life, the ink is sprayed onto papers to print 2D images or texts. The 2D printers move along the x axis and y axis, but the 3D printers move along the x and y axes with an additional z axis. The z axis enables 3D printers to print three-dimensional shape.

In order to create the final 3D models, the steps for 3D printing consist of modeling, printing, finishing. Modeling is creation of 3D printable models with software programs such as CAD and 3D modeling program. The 3D designs are converted into G-code such as Cura and Keyslicer program, which is a machine language or commands to move 3d printer parts [65]. At the printing stage, it prints a 3D model based on the 3D modeling files. There are two types of printing methods: Additive manufacturing (AM) and Subtractive manufacturing (SM). Most 3D printers use AM type, which is a method of building up a three-dimensional shape by stacking powder, liquid plastic, or plastic threads layer by layer. SM type creates a three-dimensional shape by cutting unneeded materials from a bulk material such as plastic or composites [66]. In this research, AM method was performed as the 3D printing type. Finishing is the final step of complementing the 3D printing product, painting or trimming the surface, or assembling the product parts [67].

Subtractive Manufacturing production method was generally used to manufacture the object by directly shaping the raw material of the product like a sculpture, which method is capable of precise work but takes a long time and consumes a large amount of raw material. In 1983, 3D Systems company developed the 3D printer to create three-dimensional shape by stacking the thin layers from the bottom with plastic liquid [68]. Stratasys company did the process of solidifying liquid deposited layers through a nozzle orifice, which initiated a fused deposition modeling (FDM) [69-70]. However, it has been used only for very limited application for making industrial

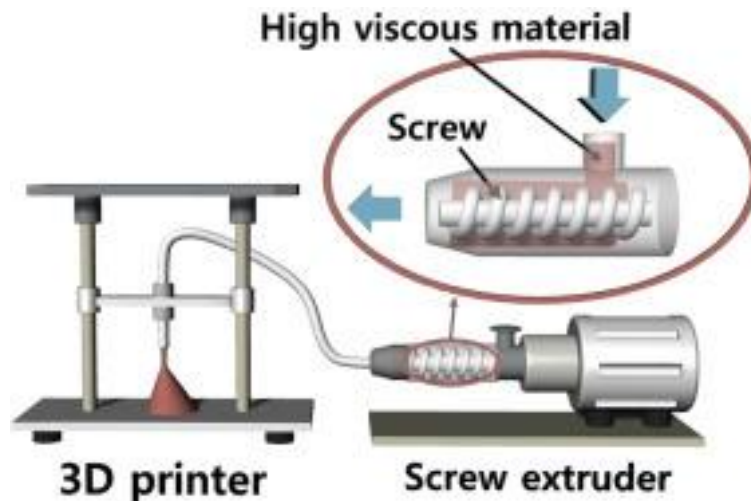
prototypes such as in the aviation and automotive industries, due to its high production costs. Studies have shown promising sustainable development by raw material reduction, ease of use and high replication rates of the 3D printers [71]. It has been developed in earnest with various applications and used in the diverse fields.

The most widely used 3D printing technology is the FDM method, which uses thermoplastic materials as a continuous filament with heating. The heated filament came out from the print extruder head and created objects by sequential layer deposition [72-73]. Not only the polymers but also metals and ceramics can be printed. A direct metal laser sintering (DMLS) is for metal printing, which laser melts the powdered metals and prints layers [74]. Binder jetting uses a binding agent with the metal particles to print metals and ceramics. Ceramic materials can be printed through extrusion and powder sintering. With the extrusion technique, the ceramics were coming out through the extruder. And the powdered ceramics are sintered with heating process in the powder sintering method [75].

## **2.2 Screw Type Extrusion (STE) 3D Printing**

A screw type extrusion (STE) mixes compounds using rotating screw extruders which are divided into single screw and multi-screw extruders. More widely use of screw extruder for plastic materials is the single screw extruders, which have low cost due to its simple structure. Multi-screw extruders have better mixing capability and higher process productivity as compared to the single screw extruders [76]. Piston type extruder can only print a certain small amount of materials. However, STE can make to print large area and continuous quantity of discharged materials by unlimited multiple feeding of the printable materials.

The Center for Printable Materials Certificate (CPMC) in The University of Texas at El Paso (UTEP) reproduced the high viscosity materials such as clay using the STE 3D printing technology and Big Rep printer with twin screw, which is described in Figure 2.1. To use clay with STE 3d printing method, water content (WC), revolutions per minute (RPM), and tip size are the variables to control stable printing speed and minimize the loss of water content during the 3D printing. Figure 2.2 is the 3D Printed enameled potteries of Ceramics using STE 3D Printing Technology [77]. STE 3D printing is generally for large scaled products like architectural applications. In recent years, 3D printing studies have been actively conducted to draw large-scaled buildings and houses using 3D printer. In 2016, Italy's company WASP decided to use clay as a printing material using a large scaled 3D printer, Big Delta, and built a large shelter at a low price [78].



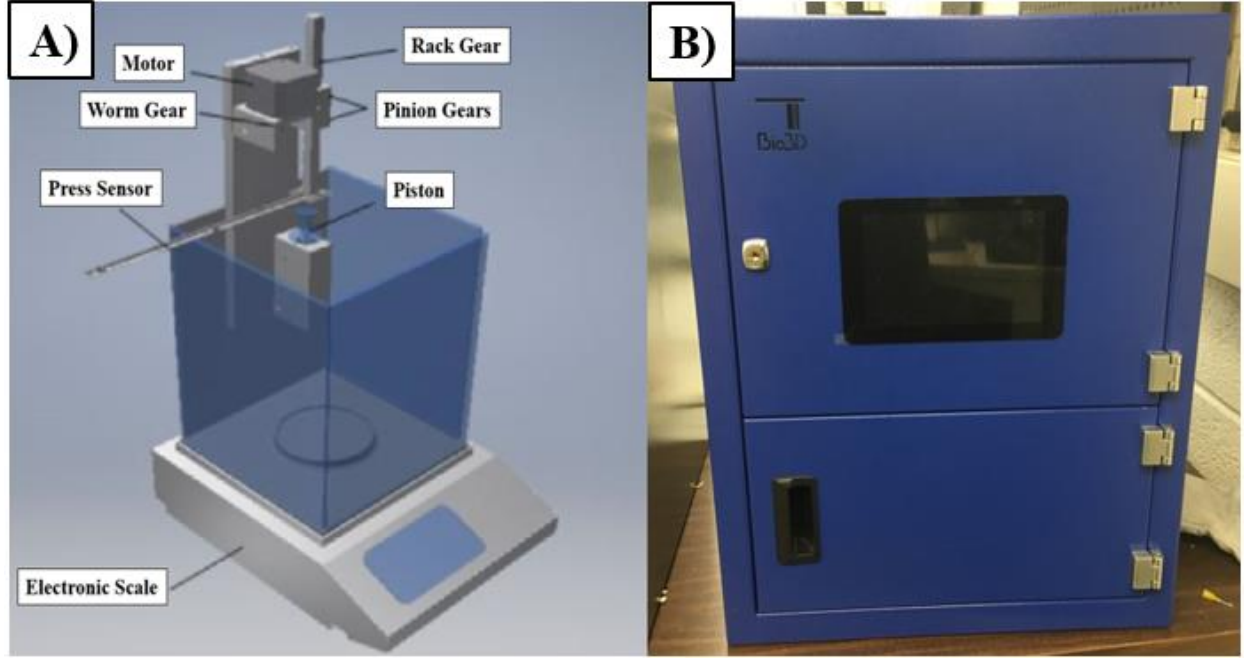
**Figure 2.1 Diagram of STE for High viscosity material and 3D Printer. [77]**



**Figure 2.2 3D Printed enameled potteries of Ceramics using STE 3D Printing Technology. [77]**

### **2.3 Piston Type Extrusion (PTE) 3D Printing**

A piston type extrusion (PTE) 3D printing is for small and precise 3D printing products. PTE printing equipment was utilized for the empirical experiments in the research, the specific images of which are shown in Figure 2.3. A and B [79]. Figure 2.3. A illustrates the conceptual diagram of PTE printing for a specially designed material. Figure 2.3. B shows the PTE method, which can print creative designs quickly with compressing the materials by mechanical pressures. PTE is capable of precision printing, but it shows significant differences depending on the forms and sizes of use [77, 80-81].

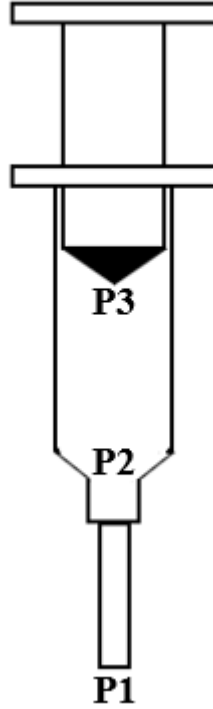


**Figure 2.3 Schematic diagram of 3D printer, which is utilized in tattoo pattern printing; A) Piston type extrusion printer (PTE), which consists of precisely controlled z-axis, IoT system, and AI technology to reproduce designs recognized by the Raspberry Pi (RPI) microprocessor. B) Actual Image of PTE Printer. [79]**

Mathematical approach for STE and PTE 3D printing methods used by Hagen-Poiseuille's (HP) equation. According to HP equation with Newtonian fluids, the amount of discharge is defined as  $Q = \frac{4\pi r^4 \Delta P}{L \eta}$ . Discharge (Q) can be increased by increasing the radius of the extruder tip (r) and increasing the pressure (P). In the Figure 2.4, The pressure at the tip  $P_1 = 1$  atmospheric pressure = 101325 Pa, the pressure between the piston and the tip  $P_2$ , the pressure at the piston  $P_3$ , the radius of the piston  $r_p = 7.3 \times 10^{-3} m$ , the radius of the tip  $r_t = 8.3 \times 10^{-4} m$ , the length of the piston  $L_p = 6.0 \times 10^{-2} m$ , the length of the tip  $L_t = 2.3 \times 10^{-2} m$ . When the total pressure is  $P_{Total} = \Delta P = P_3 - P_1 = 30000 Pa$ , the pressure  $P_3$  at the piston becomes 131325 Pa. The friction force  $\eta$  in Hagen- Poiseuille's law and material discharge from the piston ( $Q_p$ ) and the extruder tip ( $Q_t$ ) are equal. Therefore,  $Q_p = \frac{4\pi r^4 (P_3 - P_2)}{L_p}$  is equal to  $Q_t =$

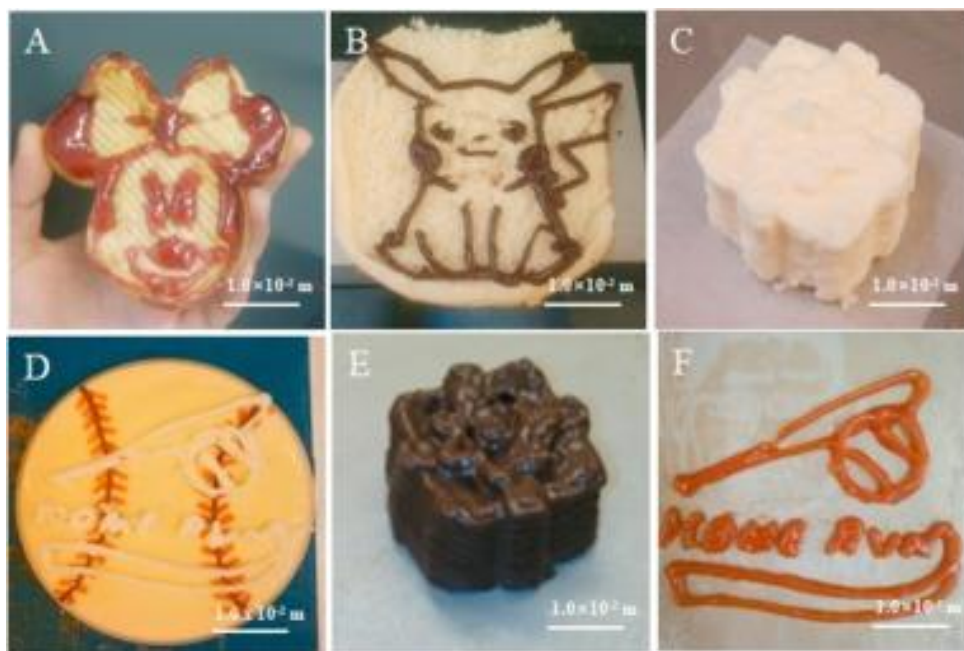


$\frac{4\pi r^4(P_2 - P_1)}{L_t}$ . It is observed that the pressure between the piston and the extruder tip and the pressure at the piston is equal,  $P_2 = P_3$ .



**Figure 2.4 Schematic diagram of Piston with Pressure P1, P2 and P3.**

Recently, the 3D printings are used in Food industry. 3D food printers draw precise shapes on coffee or prints the customized food with PTE method in refined and hygienic spaces, which describes in figure 2.5 [79]. In this thesis, the results are presented using the PTE printer and printable materials. This study brought the changes in the fields of art and science that are safe, rational, and capable of reproducibility. The manufacturing parts of tattoos such as safety and processing are controlled and supplemented, and errors in the actual fields are minimized by using 3D printers. The PTE technique is mainly applied to eyebrow, nail, and hair tattoo to maximize the effect. By printing and dying the intricate designs easily, it is possible to maintain the tattoos for a while.



**Figure 2.5 3D printed sauces onto foods using the piston type extruder (PTE): A) Jam, B) Chocolate, C) and D) Mayonnaise, E) Chocolate, and F) Ketchup. [79]**

## 2.4 Inkjet Printers for 2D and 3D printings

An inkjet printer is commonly used type of printer at home or in companies. When digital files are transferred from an inkjet printer, ink is sprayed onto paper to print 2D images or texts [82]. It is the 2D printer which moves along x-axis and y-axis. Inkjet 3D printer was developed to print with plastic and thermoplastic inks at high temperatures. The sorts of inks for the 3D Inkjet printers are water-based inks, solvent inks with volatile organic chemical compounds (VOCs), oily inks, latex inks, UV-curable inks, dye sublimation inks, and hot melt inks [83]. Even if it is not a 3D inkjet printer, 2D inkjet printer can print tattoos which do not need high temperatures to print out the materials.

## 2.5 Design patterning process

Designing the 3D printed patterns is creation of 3D printable models with 3D software programs such as CAD and 3D modeling program. The 3D designs are converted into G-code such

as Cura and Keyslicer program to command to move 3d printer parts [65]. A single line design (SLD) is a one continuous line. The SLD can easily create the designs, print fast and prevent to waste the printable materials [81, 84]. CPMC has conducted scientific and academic research about current problems for a tattoo since 2010 [57, 85-86]. Convergence technology of the tattoo process with the 3D printing method has the advantage of high precision, environmental safety, non-toxic and economic system. Furthermore, compared to permanent traditional tattoo, the novel technology using 3D printing with silver ink enables a non-permanent tattoo process, which solves the cultural conflict between youth and the old generation and realizes painless and rapid tattoo procedures.

### **2.5.1 Internet of Things (IoT) 3D Printing System**

An Internet of Things (IoT) system that controls the 3D printers based on the web was introduced with drug delivery system and telemedicine systems [85]. In order to control the IoT, the equipment that can flexibly operates the system was built on the Raspberry-pi. Users can customize the design and control in the web-based system, which system does not need to contact with the other people with non-face-to-face process. In a separate place, users can remotely draw designs through IoT control technology or send designs from photos, existing files, or web-based programs [87]. The code information based on the designs can make to start the 3D printer to print the designs. The 3D printed tattoo technology has been attempted to print the selected files onto the substrate. The 3D printing tattoo technology is expected to grow into the beauty-art field with upgraded systems and designs.

A method of 3D tattoo printing that enables remote control of IoT system is shown in the following in detail. First, it manages the system of the 3D printer equipment on the cloud and can control the operation through the web-based control from the outside of the space. The image

was chosen and transferred by the customers with cell phones or the electric devices connected over Wi-Fi as RIP or G-code file through Raspberry-Pi micro-processing, which is built into the 3D printer. The selected file is printed completely isolated from the consumer in an untact controlling system that minimizes the risks of contamination, harmful effect for the human body, and transmission of infectious diseases. It can reduce the damages caused by the process by delivering the printed results to the desired time and place for customers [88]. A built-in IoT operating system in 3D printer is the first technique applied to the tattoo in beauty-art field. Due to the simplification of the printing equipment, reproducibility, and the IoT 3D printing capable of remote control, it is considered that it can be commercialized in economic and health terms [89].

## **2.6 Printable Materials and Applications of 3D Printing**

Unlike the general 2D printers that use toner or ink as the printable materials, the 3D printers use the plastic materials with heating. As the 3D printers are used in various fields, most of the 3D printing technology applied polymers and metals as printing materials. As the printable materials become more diverse and 3D printers are increasingly developed, they are applicable to various fields such as agriculture, medical, medicine, food, manufacturing, aviation, and even education. The 3d printers can make food beyond plastic prototype, inject concrete to build houses or sculptures, and even print metal parts.

As the bioprinting technology are developed, 3D printers and bio-ceramic materials are used in the medical fields that require precision to create artificial organs such as artificial bones, joints, ears, teeth, and prosthetic arms [90-91]. The 3D bioprinting has been studied in tissue engineering applications to rebuild the complex human organs using inkjet printing. The

assistive devices or artificial bio-organs for medical use are expensive because they need to customize each product per customer. However, the cost can be lower with commercializing of the 3D printing. 3D printer company WASP (World's Advanced Saving Project) in Italy printed 3D medical products such as cranial, bone implant and prosthetic legs. They modeled 3D scanned orthosis using a free add-on for modeling 3D medical products [92].

The 3D printing is used in construction fields. ETH zurich built nine concrete columns without formwork in Switzerland using a large scaled 3D printer. They used a layered extrusion printing type to produce the large concrete architecture: column height 2.7m; print speed 180 mm/sec; time per column 2.5 hours [93]. The 3D technology is also needed in clothing fields. 3D print clothing achieved various complex designs which would be impossible to construct by traditional clothing method [94]. Nike and New Balance made shoes with 3D printers for football athletes and runners [95].

There is an example that 3D printing has also been used in education. Sierra Leone's Bio President created a 3D print of the distribution of the number of girls who do not attend major schools around the city, and printed difficult educational data using the 3D printing. Through the 3D printed creature, many people were able to see the data from different angles at once, and able to help understanding with having conversation each other. Furthermore, the printed data could be discussed without a battery or electricity [96]. The 3D printing is used in beauty-art field. IoT controlled- 3D nail printing did makeup on nail with designs, which are described in Figure 2.6 [97].



**Figure 2.6 Printed nail shapes by IoT controlled 3D Printers. [97]**

In recent days, the 3D printing began to be used in the food industry using edible ingredients as printable materials including chocolate, ketchup, or peanut butter in a refined environment [98]. A 3D food printer Foodini printed food such as pizza and pasta with fresh ingredients [99]. NASA is also studying for the 3D food printing technology to prevent food waste and to make customized food for astronauts [100]. As mentioned before (at the 2.3 Piston Type Extrusion 3D Printing section), 3D food printers draw precise shapes on the food based on demand with PTE method in a clean space.

### **Chapter 3: Materials and Experiment**

Various concentration of non-permanent silver tattoo ink was produced, and the dyeing preservation period was varied with different silver contents. Non-permanent tattoo procedure involves producing hygienic silver ink patch, which is patterned as tattoo draft and solidification; pre-treatment process on human skin to provide moisture and oil through utilizing microneedle; final dyeing process by putting patterned patch which contains hygienic silver compound on the skin layer. The silver ink tattoo is designed to spread silver and dye stratum corneum layer of human skin, and the tattoo could last 3 days to 2 weeks (supposed that the client's skin has proper nourishment and moisture). After specific days, the tattoo pattern could fade away and disappeared with dyed skin cells. The tattoo experiment was conducted on a different part of the human skin and nail.

#### **3.1 Synthesis of Silver Nitrate Ink**

Non-permanent tattoo ink was produced with the primary material of silver, and one of the silver compounds (among Ago, AgO<sub>2</sub>, Ag<sub>2</sub>O, AgNO<sub>3</sub>, AgBr, AgCl, and AgI) was utilized in the process. The weight ratio between solvent (oil or distilled water) and solute (silver compound) was controlled by the dyeing preservation period on the stratum corneum layer. The dispersing agent which assists viscosity of tattoo ink was selected as polyvinyl alcohol (PVA), polyethylene glycol, ethylene glycol, polyvinyl pyrrolidone (PVP), cyclodextrin, sodium oleate, valine, octadecylamine and dodecanethiol (all the chemicals were purchased from Sigma-Aldrich, Missouri, MO, USA). Various concentration (2, 5, and 10wt% of silver compounds) of the silver solution was produced with distilled water.

Gelatin hydrogel was prepared to absorb silver particles on the pattern surface. Gelatin hydrogel is designed to be printable by utilizing piston-type extrusion (PTE) printing technology.

After printing, the patterned substrate would soak in silver solution to produce a silver-containing patch. 0.5g of gelatin was utilized, and 0.5g of PVP was used to assist the solidification of a pattern. Solidification was conducted in the freezer under -5 °C for 30 minutes. And also the collagen-based microneedle can be the substrate. After freezing or drying the substrates, the substrates were dipped into the silver nitrate tattoo ink.

### **3.2 Contact Angles of Silver Nitrate Inks**

Contact angle of the materials can be measured depending on whether each material is hydrophobic or hydrophilic material. If the contact angle of the material is more than 90 degrees, this material is classified as a hydrophobic surface and if it is below 90 degrees, it has a hydrophilic surface [101]. Contact angle is extremely sensitive by contaminations, so the solid surface needs to be clean. To measure the contact angle, contact angle goniometer (Raméhart instrument co.) and DROPImage Advanced were used. A drop of each 2%, 5% and 10% silver nitrate inks of the micro-syringe was released onto a metal surface and then their contact angle was measured. The software measures contact angle, height and width of the drops and capture the images of the droplets.

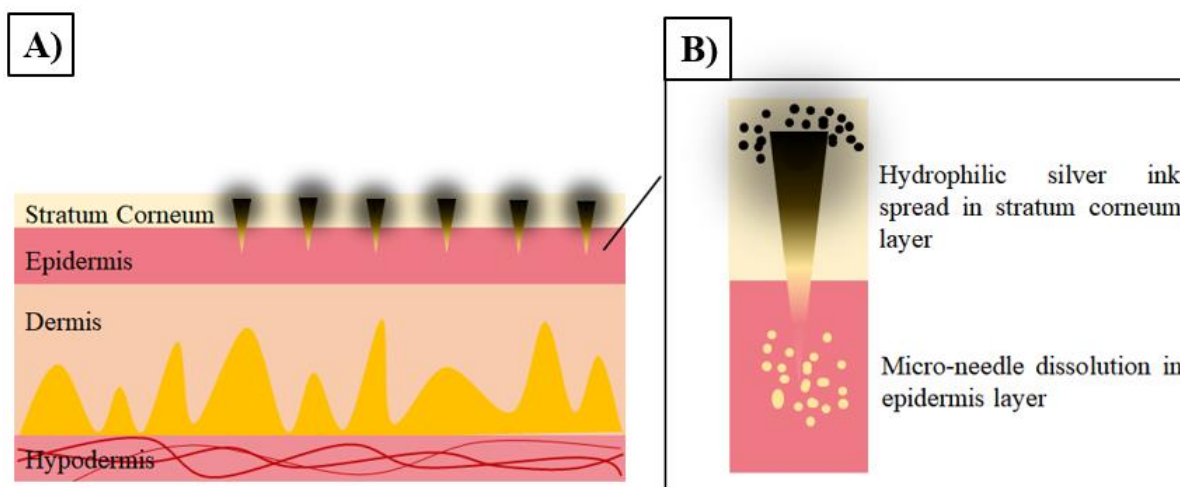
### **3.3 Production of Microneedle Patch for Non-permanent Tattoo**

At the 52nd Winter Conference of the Korean Society of Cosmetology in December 2019, Namsoo Peter Kim introduced a method of injecting tattoo inks into the skin by making microneedles with collagen biomaterials. It is possible to do non-permanent tattoos using 3D printer without needles nor anesthetics [102]. New techniques have been utilized to control the diffusion rates of tattoo inks with microneedles and, in particular, to prevent the ink from penetrating the dermis layer of skin. The microneedle patch can control the quality of non-



permanent tattoo and also the accurate dispersing speed and depth of tattoo ink. The microneedle could be injected before putting the silver ink patch on the skin.

As shown in the Illustration 3.1, a hydrophobic property of a collagen-based needle penetrates up to the epidermis layer, which is 300-600 $\mu\text{m}$  in depth. At the stratum corneum layer, hydrophilic silver ink spreads only in a specific area, and the dispersing speed is controlled by microneedle. On the surface, the hydrophilicity and hydrophobicity were improved to combine with the surroundings of the needle. It makes to slow the ink diffusion rate into the skin, allowing the silver tattoo ink to stay within a given period.



**Illustration 3.1 Schematic images of silver ink dispersing on the skin layer with a microneedle.** A) Microneedle with silver ink on the skin layer. B) Silver inks disperse in the epidermis layer.

Since the stratum corneum is usually replaced periodically with a cycle of 2 weeks, the printed tattoo could disappear naturally when a new stratum corneum is created. The experiment is based on a method in which the tattoo ink does not diffuse into the epidermis and dermis layers within a given time and stably remains in the stratum corneum. According to Fick's 2nd law, the ink concentration and diffusion rate are controlled by the penetration rates of keratin in the stratum

corneum that is not absorbed into the dermis. Ultimately, the experiment was designed to release the ink along with the cycles of the stratum corneum.

Different from the existing tattoo, the users select the tattoo existing period, and the microneedle could be injected to change the stratum corneum into hydrophilic to control the ink diffusion rates. This provides a disposable tattoo by discoloring the tip of the needle. Also, this can be used to improve the moisturizing and regeneration of the skin with hyaluronic acid or vitamins. It can prevent the destruction of the components of the solution containing hyaluronic acid and vitamins because the heat treatment is not performed during the dipping process for the nutrients.

Also, microneedles with a thickness less than  $5\mu\text{m}$  do not make contacting with the dermal layer, so it is possible to produce mild cosmetics that help moisturizing and regeneration of skins. However, it could not be used in this study because the ink maintaining time and collagen should not act as variables for controlling the ink diffusion rates. By adjusting the concentration 1~100,000 cp, the clients can easily do the tattoos as various patterns and colors.

### **3.4 Printing Tattoo with 3D Print Technology**

The disposable and non-permanent tattoo is printed by the PTE printers and solidifies. The tattoo reacted and decolorized in the stratum corneum with the pattern of the silver compounds tattoo ink by attaching the solidified features to the skin. There are many differences in printing and control of ink diffusion rates on skin surface depending on people, body parts, contact time, temperature, moisture, and oil contents.

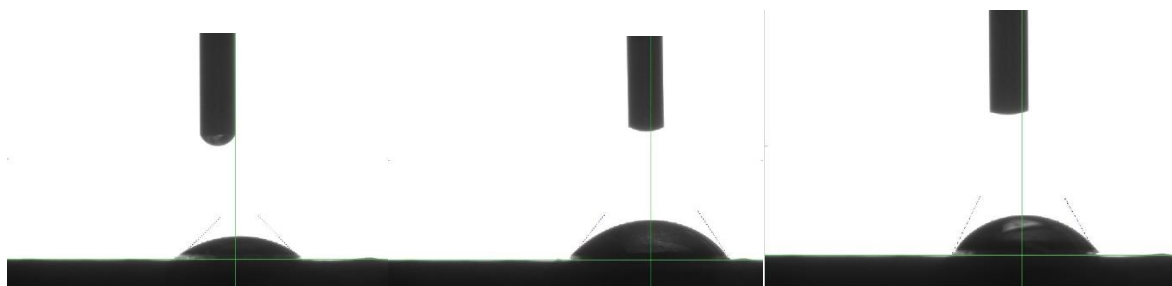
Since these experiments have a combined effect of several external and internal variables, the concepts were first introduced in South Korea in 2010 [86]. Empirical experiments were

conducted with a focus on the stability and safety of the materials based on thousands of test results over 10 years. The experiments focused on the nails and the back of the hands. Tattoos were printed on the body parts based on the results of lasting optimization experiments according to the permeability of the stratum corneum, which depends on the concentrations of silver compounds. Besides, a new concept of a non-permanent tattoo was attempted to diffuse 10-100 $\mu$ m thick from the skin surface. The simulation results of the ink diffusion distances in the keratin stratum corneum of the skin were obtained by the concentration and time variables. In consideration of the stability, the non-permanent tattoo was carried out according to the diffusion distance simulation.

## Chapter 4: Results

### 4.1 Contact Angles of Silver Nitrate Inks

To figure out if the substance is hydrophilic or hydrophobic with various concentration of the silver nitrate inks, the contact angles were measured. A droplet of each 2%, 5%, and 10% silver nitrate tattoo inks is released onto the substrate, which is described in the Figure 4.1. And Table 4.1 indicates the results of mean values of angles, heights and widths of 2%, 5% and 10% Silver nitrate inks. The mean values of the angles were  $45.93^\circ$  (for 2%),  $54.43^\circ$  (for 5%), and  $62.83^\circ$  (for 10% concentration of the silver nitrate ink). The results describe that the silver nitrate ink has hydrophilic properties, and has hydrophobic characteristics with increasing the concentration of the silver nitrate ink.



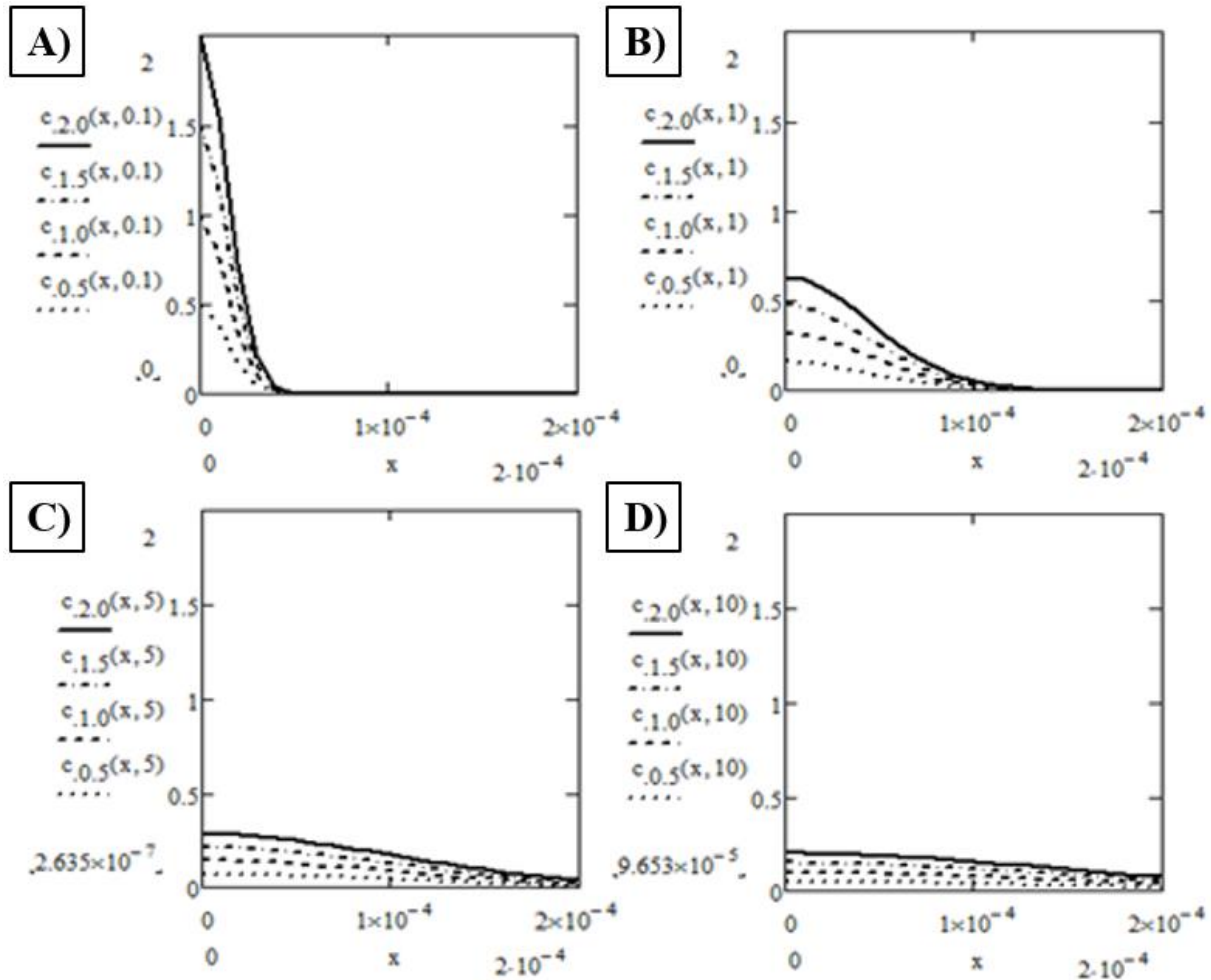
**Figure 4.1** The images of the droplets of 2%, 5% and 10% Silver nitrate ink.

**Table 4.1 Results of mean of angles, heights and widths of 2%, 5% and 10% Silver nitrate inks.**

No	Silver nitrate inks								
	2%			5%			10%		
	Mean of Angle (°)	Height	Width	Mean of Angle (°)	Height	Width	Mean of Angle (°)	Height	Width
1	46.3	0.431	2.324	54.8	0.756	3.032	63.3	0.692	2.441
2	46.1	0.430	2.324	54.7	0.755	3.033	63.1	0.691	2.446
3	46.1	0.429	2.323	54.7	0.755	3.032	63.0	0.689	2.448
4	45.9	0.427	2.323	54.6	0.753	3.033	62.9	0.686	2.449
5	45.9	0.426	2.323	54.5	0.750	3.033	62.9	0.684	2.451
6	45.8	0.425	2.323	54.4	0.749	3.033	62.8	0.682	2.451
7	45.8	0.424	2.321	54.3	0.747	3.033	62.7	0.681	2.451
8	45.9	0.423	2.318	54.1	0.746	3.033	62.6	0.680	2.452
9	45.8	0.423	2.319	54.2	0.744	3.031	62.5	0.680	2.452
10	45.7	0.422	2.318	54.0	0.743	3.032	62.5	0.679	2.452
Average	45.93	0.426	2.3216	54.43	0.7498	3.0325	62.83	0.6844	2.4494

#### 4.2 Diffusion Rates

The simulation of diffusion of silver ink in skins using PTE method was explained by Fick's 2nd law  $\frac{\partial c}{\partial t} = -D \frac{\partial^2 c}{\partial x^2}$ , which describes the diffusion rates of the silver tattoo ink depends on the concentrations of ink (0.5, 1, 1.5 and 2 wt%) and the distance in skins (see Figure 4.2). The data expect diffusion rates depending on the exact penetrated distance of the ink in skins using the different concentrations of the silver ink after the contact time. Figure 4.2 represents the results of the Distance-concentration graph at contact time 0.1, 1, 5, and 10 minutes. Different concentrations of silver ink were sprayed on the skin to evaluate the penetration rates into the skin. The amount of silver ink decreases as the skin gets more rooted within the stratum corneum based on the graph.



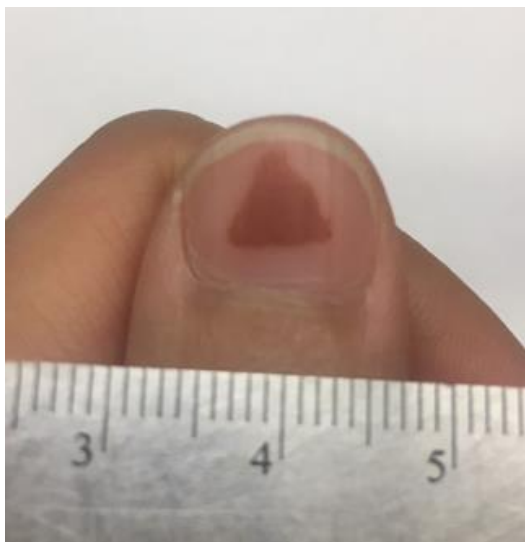
**Figure 4.2** The graphs of the diffusion rates of the silver tattoo ink with different concentrations of ink (0.5, 1, 1.5 and 2 wt%), contact time (minutes), and the distance (m) in skins. The representative results show the different rates of the silver tattoo ink under different conditions; Distance-concentration graphs: A) at contact time 0.1 min, B) at contact time 1 min, C) at contact time 5 mins, and D) at contact time 10 mins.

Projected figures were successfully removed following the calculated penetration rates. The dispersing speed rate controls the duration period of projected shape on the stratum corneum and removing speed. Based on the result in the Figure 4.2, the proper contact time and the concentration of the silver nitrate ink are figured out. The slope of the graph should be nearly constant to diffuse ink slowly and stay in the given skin cycle. Figure 4D with 10 minutes of the contact time is the most ideal graph to diffuse and hold the ink concentration in the skin cycle.

Depending on the moisture and oil contents of different locations of skin, thicknesses, and temperature, different printing conditions were applied to achieve specific diffusion rate and duration period. The micro collagen needles were utilized to make the similar contents of moisture and oil for a different type of skin. Nutritious oil and moisture were supplied before 30 minutes of a tattoo. The limitation of permanent tattoos and henna proved the effectiveness of the non-permanent tattoo technique. Dyeing of the stratum corneum by the silver ink compound with disposable microneedles provides a hygiene and economic tattoo process.

#### **4.3 2D/ 3D Printing Patterns with PTE 3D Printing Method**

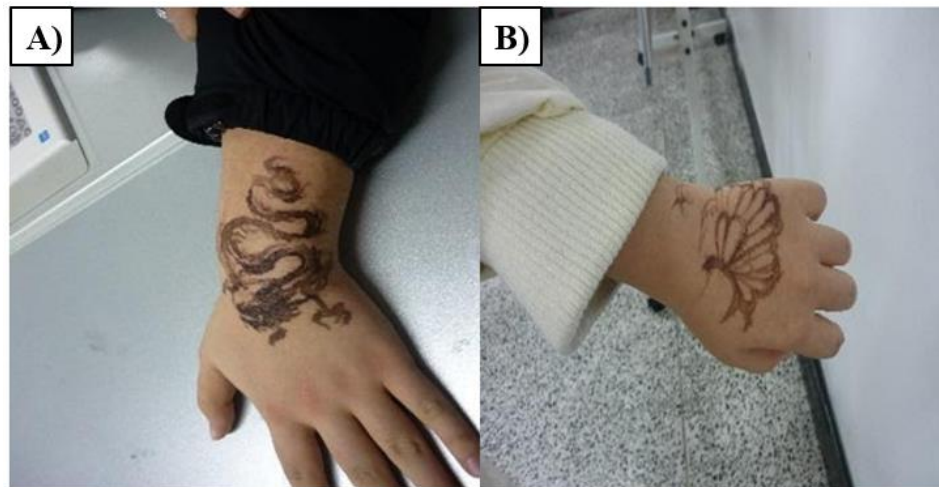
As shown in Figure 4.3, silver ink was projected on the nail by consideration of the penetration depth and the concentration of tattoo inks with the PTE method. The results were achieved by using 2wt% silver ink for 5 minutes. The pigment on the nail was stable and maintained the figure until a new nail grew, and the figure faded away after 10 days.



**Figure 4.3 Image of the stably projected figure on the nail using 2 wt% of silver ink after 5 minutes. The Triangle pattern was formed by the PTE method.**

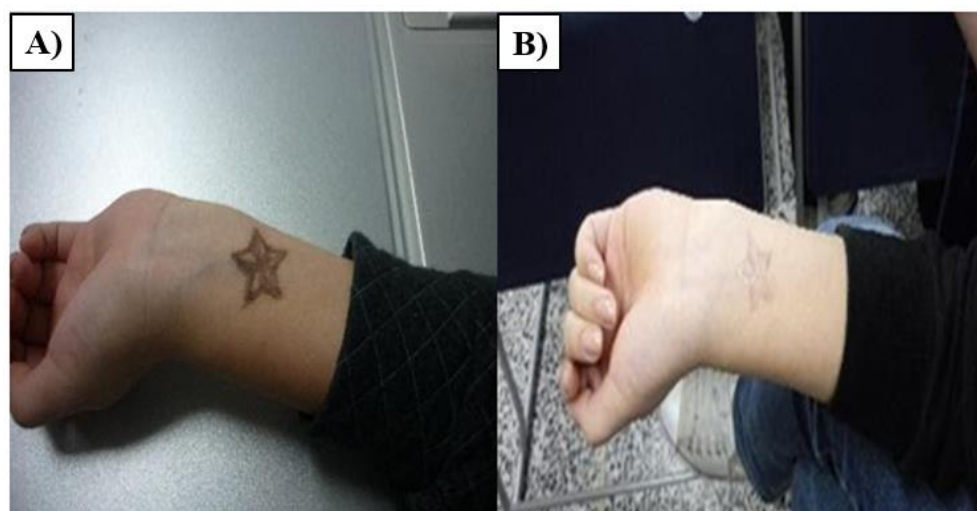
Based on the data of penetration rate and concentration, the non-permanent tattoo was conducted using a dragon-designed draft on hand (Described in Figure 4.4). After providing a

proper amount of moisture and oil on hand, patterned patch with 0.5wt% of silver compounds ink dyed skin for 10 minutes. Despite washing with water, the dragon figure on hand had lasted for 11 days. However, keratin in the stratum corneum should be eliminated using force in specific cases like tattooing on parts where there is much contact.



**Figure 4.4 Non-permanent tattoos applied on hands using 0.5wt% of silver nitrate ink for 10 minutes. A) Dragon; B) Butterfly.**

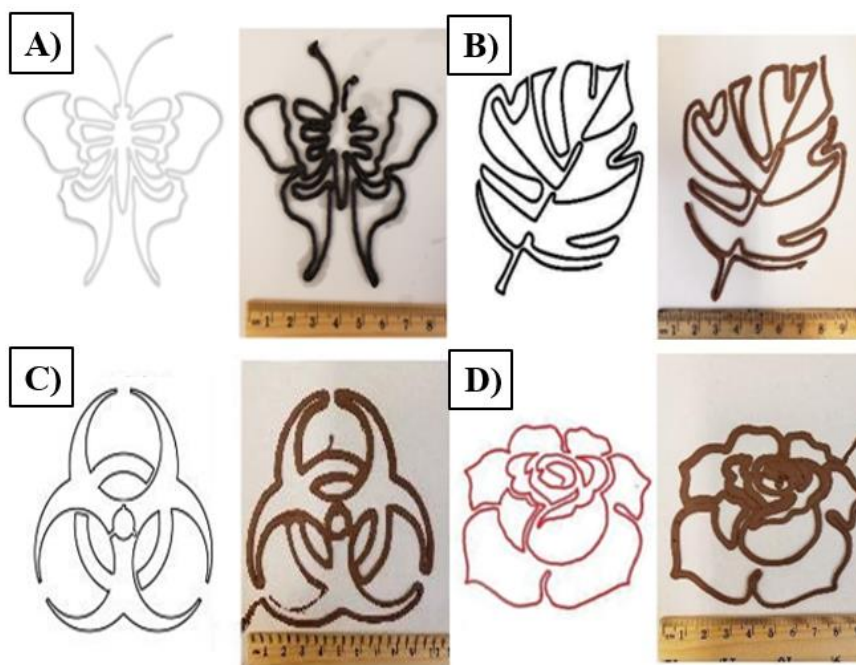
As Described in Figure 4.5, the duration of a non-permanent tattoo was proved that the star tattoo disappears after 7 days with 0.2wt% concentration of tattoo ink. Regarding the several of parameters such as projection place and the cell cycle of the stratum corneum, the tattoo removal rate following silver concentration could be predictable.





**Figure 4.5 Images of a non-permanent star tattoo applied on human skin using 0.2wt% of silver nitrate ink. A) After the tattoo experiment; B) After 7 days.**

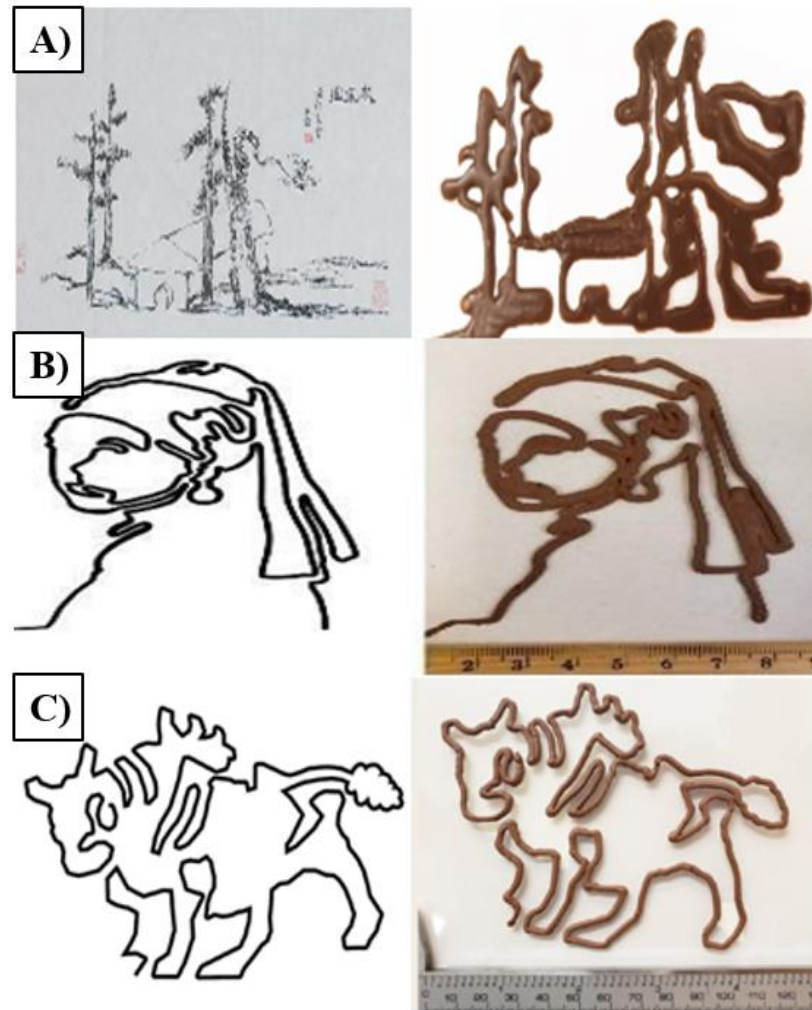
The experiment result evaluated the efficacy of non-permanent tattoo using a silver compound, and this new process has flexibility for cosmetic applications; analgesics, antibiotics, disinfectants, and aesthetic expressions of the wounds through the control of drugs and ingredients. Figure 4.6 shows the applications of 3D printing in art, which are usable for tattoo draft fields. According to Figure 4.6, PTE printing method makes available of forming delicate images using hydrophilic ceramic material.



**Figure 4.6 Images of a pattern using hydrophilic ceramic material. A) Butterfly; B) Leaf; C) Emblem; D) Rose.**

From a different perspective, Figure 4.7 shows the artistic applications of 3D printing, which describes imitative 3D printed draft of masterpieces; Winter scene by Jeong-hui Kim; The girl with a pearl earring by Johannes Vermeer; White cow by Joong-shop Lee. The printed

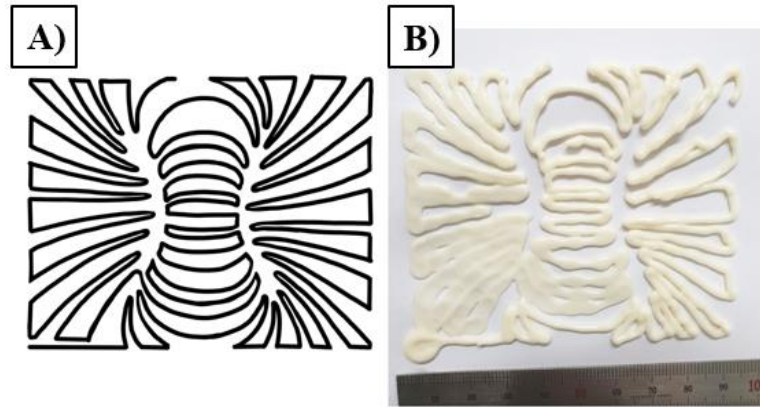
applications preserved the characteristics of the existing works of the masterpieces, but was simply designed by single line design (SLD) and printed quickly.



**Figure 4.7 Images of a pattern using hydrophilic ceramic material.** A) Winter scene; B) The girl with a pearl earring; C) White cow.

The optical illusion techniques can be applied to two-dimensional designs. In Figure 4.8, SLD designs look like a complex three-dimensional form with the optical illusion technique, although it is a two-dimensional form. The effect of the illusion technique can be maximized when moving the printed tattoo on the eyebrows and hairs. It has successfully demonstrated that non-professionals of art can enter a field of art, and there is an advantage that can be extended the artworks to the non-permanent tattoos. Printing creations and expressing them on the part of the

human body could be applied to a beauty-art industry. The research reinterprets the existing artworks and successfully applies them to the non-permanent tattoo fields.



**Figure 4.8 Images of a pattern by the optical illusion technique.** A) Image of optical illusion design; B) Image of a pattern using the PTE method.

## **Chapter 5: Discussion**

### **5.1 Contact Angles**

In the experiment, 2%, 5% and 10% silver nitrate inks were used to measure the contact angles. According to the results of the contact angles, the contact angles increased with increasing concentrations of the solution (2% to 10%). As it mentioned before, hydrophilic substrate means when contact angle is smaller than  $90^\circ$ , and hydrophobic substrate means when contact angle is bigger than  $90^\circ$ . The contact angles of 2%, 5%, and 10% silver nitrate were  $45.93^\circ$ ,  $54.43^\circ$  and  $62.83^\circ$ , respectively. Those are hydrophilic materials with contact angles less than  $90^\circ$ . With adding more amounts of silver nitrate in solution, the solution becomes more hydrophobic characteristics. Therefore, elimination of sources of error and contaminations is vital to produce reliable results. The contact angle goniometer is highly recommended to measure the optimized result of the contact angle and to see actual image of the drop.

### **5.2 Future Applications using Tattoo 3D Printing Technology**

With the spread of COVID-19, the population all over world is suffering from the corona virus. A non-face-to-face contact program was needed depending on the situation where people could not meet face-to-face, work, or eat. The pandemic has had serious negative impacts on both individual's health and global economies. There was opportunities and challenges for tattoo technology. The extreme situation enables IoT-3D printing technology to commercial or developed. Also it can reduce the hazards from the lack of proficiency and hygienic problem of tattooists by using precise design and control of the 3D tattoo printing technology in the tattoo fields.

The IoT-3D printing technology with the micro-needle patches will grow into beauty-art fields for treatment as pharmaceutical uses and cosmetic purposes with some improvements. This

is because the microneedles are consisted with collagen which have the advantages of biodegradability, biocompatibility and non-toxic properties. The collagen can reinforce the bones, teeth and skins.

### **5.2.1 Adding Ingredients: Vitamins or Hyaluronic acid**

In 2018, the drug delivery system with the 3D printing technology was successfully performed. Quantitative drug dosages were mixed into the composition of food and printed along the 3D designs for health care. The PTE 3D printer printed the quantitative dosages of vitamin C which were melted in water, ketchup, mayonnaise, and peanut butter [85]. Based on the result of the experiment, the microneedle silver nitrate tattoo patch can be used to improve the moisturizing and nourishing the skin with adding ingredients such as vitamins and hyaluronic acid. Vitamins and hyaluronic acid are sensitive at elevated temperatures. However, the nutrients cannot be destructed their molecular structures in the collagen microneedle. This is because there is no heat treatment process to manufacture the patch.

In the study, the silver nitrate tattoo technology was not possible to combine the vitamins or hyaluronic acid with microneedle. Because of some factors such as the time that ink maintains and the concentration of the silver nitrate, they can affect the result of the diffusion rates and remaining times of the tattoo ink experiment. In the cosmetic field, further research can be fulfilled with adding the nutrients into the collagen microneedle patch without or with silver nitrate ink tattoo by adjusting the factors.

### **5.2.2 Artistic Application of 3D Printing on Beauty-Art**

The 3D silver nitrate ink printing technology can be applied to the beauty-art such as nail, hair, and eyebrow tattoos. The 3D tattoo printing can be done and be extended the fields by non-professional persons with ease of use. Compared to conventional procedure, the procedure of non-permanent tattoo with 3D printing technology on the nail, hair, or eyebrow takes a little time. With the SLD design, the 2D design looks like a complicated 3D form. The optical illusion techniques can be maximized by shaking or moving the printed tattoos on hairy parts.

## **Chapter 6: Conclusion**

It was possible to produce recognizable shapes based on design programs using the silver nitrate tattoo ink without interference from the printability issues that commonly arise when printing with high viscosity materials. Experiment results proved that the PTE 3D tattoo printing method has advantages of the structural formation using various ingredient-controlled tattoo ink.

Furthermore, 3D printed tattoo materials have a fundamental strength for the transferring tattoo design because of porous materials and surface tension to control the tattoo ink into the skin. The data collecting is essential to be done to construct more database for printing conditions using other tattoo ink materials and to prove that 3D methodology. This research will lead to many possibilities and interests in the beauty-art industry and provide ideas for further study in new tattoo methodology and 3D printing application of tattoo production.

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## **Vita**

The author of the thesis, Jisu Lee was born in Seoul, South Korea, on August 21st in 1996. She studied in kindergarten, elementary school, middle school and high school while living in Seoul. After graduation of the high school, she entered Seo-Kyung University (Seoul, South Korea) on March 2015. She studied for a Bachelor's degree in Nano Convergence Engineering in SK University. On August 2017, she transferred to University of Texas at El Paso (UTEP) to study for the Bachelor's degree in Metallurgy, Materials and Biomedical Engineering (MMBME). On Spring 2018, she was elected for the University of Texas at El Paso's Dean's list. After graduation of the Bachelor's degree in MMBME, she started to study for a Master's degree in MMBME. She then started to research about 3D Printing technology at the Center of Printing Materials Certificate (CPMC) as a graduate research assistant (RA). After that she helped Dr. Kim's class (Rate Process) as a graduate teaching assistant (TA). She researched about biomaterials such as extracellular matrix (ECM) and high viscosity 3D Printing Technology with Ceramic materials, screw extruder type printer (STE), and Big rep printer. She then also researched about non-permanent 3D printing tattoo using silver nitrate ink. The author defended the thesis on Thursday, October 29th, 2020 at 5PM (Time in El Paso).

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