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Metallic Nanoparticles Inks for 3D Printing of Electronics

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The 3-dimensional (3D) printed electronics sector in the additive manufacturing (AM) industry has grown substantially for the past few years, and there is increasing demand for different types of metallic nanoparticles inks in electronics printing for various applications. Metallic nanoparticles inks are commonly used for fabricating conductive tracks and patterns due to their relatively higher electrical conductivity as compared to other types of inks, and they can be further categorized into single element metallic nanoparticles inks, alloy metallic nanoparticles inks, metallic oxides nanoparticles inks and core-shell bimetallic nanoparticles inks. It is critical to gain a deep understanding of the metallic nanoparticles inks used in 3D printed electronics as the material properties of these inks can directly affect the final electrical and mechanical properties of the printed patterns. This review presents an overview of the available metallic nanoparticles inks used for 3D printing of electronics, and critically reviews the strengths and weaknesses for each type of ink. Finally, the challenges of metallic nanoparticles inks in 3D printed electronics are also discussed along with the future outlook of 3D printed electronics.

1. Introduction

3-dimensional (3D) printed electronics have captured much attention in the recent years, owing to their success in allowing on-demand fabrication of highly-customizable electronics on wide varieties of substrates (for instance, polymer films, papers and textiles) and conformal surfaces. 3D printing of electronics is an interdisciplinary field which integrates materials science with engineering principles, in which machines precisely deposit conductive inks onto substrates (for instance, direct-writable printing technologies such as aerosol jet and inkjet printers [1]). Current conventional electronics fabrication methods comprise of a series of additive and subtractive manufacturing techniques which are complicated, time-consuming, uneconomical and environmentally unfriendly. Therefore, 3D printing of electronics aims to mitigate these shortfalls and at the same time, allowing low-cost fabrication of electronics and helps to increase designs freedom by allowing electrical circuits and components to be directly fabricated onto various types of substrates [2-14].

Although 3D printed electronics has the potentials to allow low-cost rapid prototyping of functional electronics on different substrates for various applications, the inks used in 3D printing of electronics are hindering the progression of the 3D printed electronics due to their high price tags and low electrical performances. The inks are one of the most critical components in the 3D printing of electronics, in which the final electrical and mechanical properties of the printed patterns are affected by the material properties of the inks. Therefore, it is critical to gain a deep understanding of inks used in 3D printed electronics. Different types of inks have different specialized properties and functionalities, and some of the available inks in the market are metallic nanoparticles inks, metallic-organic decomposition (MOD) inks, conductive polymers inks, dielectric inks, semiconductor inks, carbon nanotubes inks and graphene inks (see **Figure 1**).

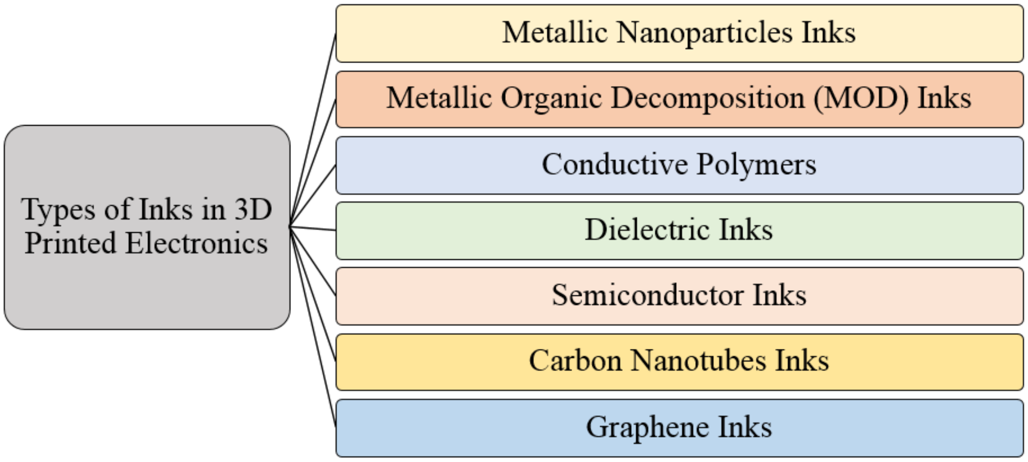

Figure 1. Different types of inks used in 3D printed electronics.

Metallic nanoparticles inks are readily obtainable from the commercial market and are commonly used for fabricating conductive tracks and patterns due to their relatively higher electrical conductivity as compared to other types of inks. These metallic nanoparticles inks are made up of electrically conductive metallic nanoparticles suspended in liquid mediums, with particle size ranging from 1 – 100 nm. However, there are also diverse types of metallic nanoparticles inks in the market (for instance, silver nanoparticles inks, gold nanoparticles inks and copper nanoparticles inks). Metallic nanoparticles inks of different material compositions can directly influence the final electrical, material and mechanical properties of the printed patterns. This paper presents an overview of the different types of metallic nanoparticles inks used for the 3D printing of electronics. The strengths and weaknesses of each type of ink are critically reviewed. The challenges and potentials of metallic nanoparticles inks in 3D printed electronics are also discussed.

2. Metallic Nanoparticles Inks

Metallic nanoparticles inks are suspensions of metallic nanoparticles in liquid mediums (see **Figure 2**a), in which individual metallic nanoparticle is encapsulated in a layer of insulating organic additives and stabilizing agents (see Figure 2b). The encapsulating organic additives and stabilising agents help prevent the metallic nanoparticles from agglomeration, but at the same time, also impede the flow of electrons from particles to particles. Therefore, sintering processes are required to remove these liquid mediums and encapsulating organic additives from the wet as-deposited printed patterns so that the metallic nanoparticles can form contacting points with neighbouring particles to conduct electricity (see Figure 2c). The printed patterns mainly consist of metallic nanoparticles after the decomposition of liquid mediums and organic additives. Therefore, the material composition of the metallic nanoparticles largely determines the electrical, mechanical and material properties of the printed patterns.

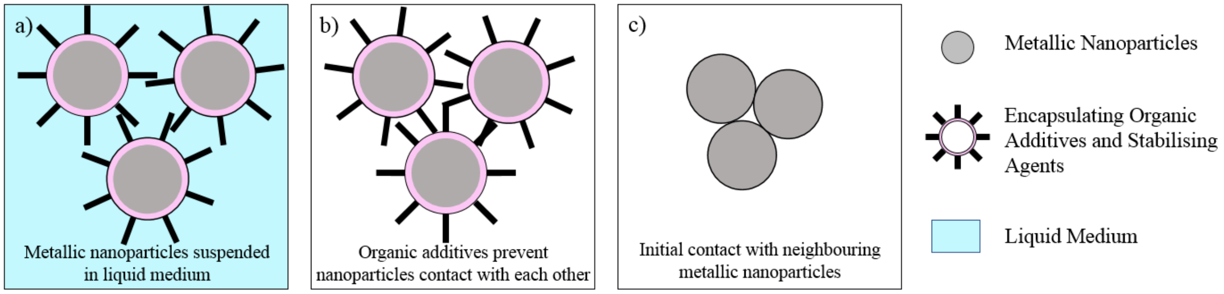

Figure 2. a) Metallic nanoparticles suspended in a liquid medium, b) organic additives prevent nanoparticles contact with each other, and c) initial contact with neighbouring metallic nanoparticles.

2.1. Parameters of Metallic Nanoparticles

Although the electrical, material and mechanical properties of the printed patterns and the required sintering temperatures are primarily influenced by the material compositions of metallic nanoparticles inks, other parameters can also affect these properties in one way or another. The parameters of metallic nanoparticles inks can be categorized into metallic nanoparticles and liquid medium (see **Figure 3**). Metallic nanoparticles can be further classified according to their material compositions, particle size, particle shape and particle concentration; and liquid medium can also be further classified according to the carriers, dispersants and additives [15]. The particle size can impact the melting point of the metallic nanoparticles, in which nanoparticles have significantly lower melting points than their bulk counterparts. For instance, 5nm diameter spherical gold nanoparticles have a melting temperature of less than 300 °C as compared to bulk gold's melting temperature of 1064 °C [16-20]. This unique phenomenon allows the sintering of nanoparticles at a much lower temperature [18-20]. In addition, the particle shape may also influence the optical, electrical, magnetic and catalytic properties of metallic nanoparticles [21-22]. The solid loading of the metallic nanoparticles inks can also influence the electrical conductivity of the printed patterns directly [23-24]. Furthermore, the decomposition temperatures of the organic additives and stabilizers in the inks may also affect the required sintering temperatures [21-22], as these organic additives and stabilizers are required to be removed before coalescence of the metallic nanoparticles can take place.

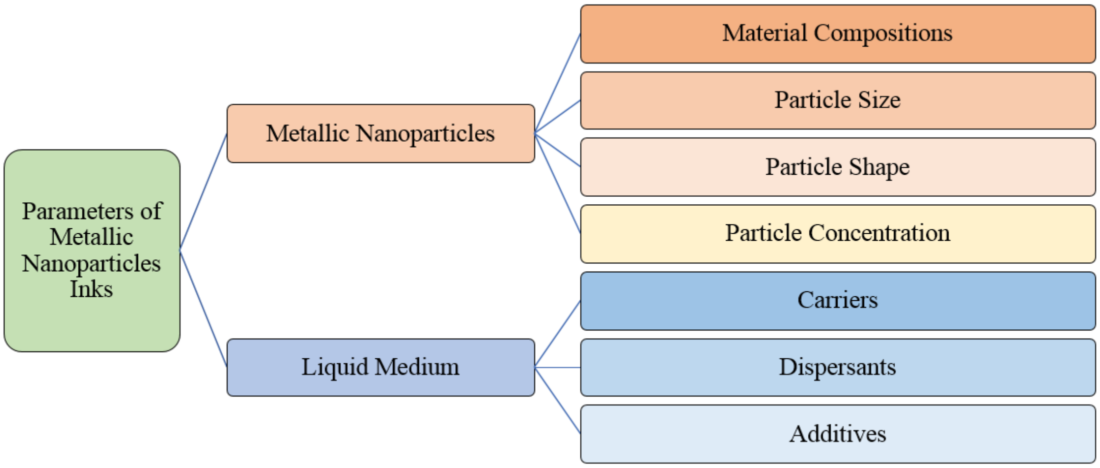

Figure 3. Parameters of metallic nanoparticles inks [15].

2.2. Choice of Metallic Nanoparticles

There are several important factors to consider for deciding the choice of metallic nanoparticles for the ink: electrical conductivity, oxidation stability, costs, and the desired electrical and magnetic properties (see **Figure 4**). However, individual application's requirements primarily determine these factors. 3D printed electronics applications generally require the printed conductive tracks to have minimum electrical resistance to reduce Joule heating effects and promote efficiency.

Therefore, it is logical to choose materials with better electrical conductivity as a choice of material composition for metallic nanoparticles inks. For instance, silver, gold and copper are usually preferred as a material choice for metallic nanoparticles inks due to their superior electrical conductivity. Apart from electrical conductivity, the material composition of metallic nanoparticles should also possess good oxidation stability. At elevated temperatures, some metals tend to form metal oxides easily when exposed to air and these types of metals are not ideal for formulating metallic nanoparticles inks. For instance, copper nanoparticles rapidly oxidize in the atmosphere at elevated temperatures, and these undesirable copper oxides are found to be significantly less conductive than pure copper nanoparticles [19]. Although good electrical and material properties are crucial, their material costs cannot be neglected too. As 3D printed electronics aim to decrease fabrication costs, the material costs must not be too high. For example, highly conductive gold nanoparticles inks exhibit excellent electrical properties and oxidation stability, but the price is too high for cost-effectiveness in mass productions of cheap 3D printed electronics. Therefore, ongoing research has been conducted to search for suitable materials for formulating metallic nanoparticles inks that possess good electrical and material properties with significant low cost. Finally, materials with unique electrical, material and magnetic properties may also be a factor of consideration for the choice of material composition.

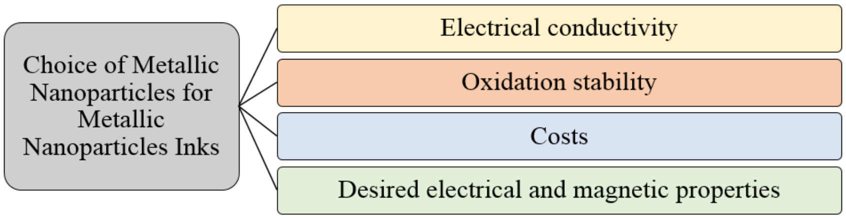

Figure 4. Choice of metallic nanoparticles for metallic nanoparticles inks.

2.3.Types of Metallic Nanoparticles Inks

These metallic nanoparticles inks can be further categorized according to the material compositions of the metallic nanoparticles. Hence, the metallic nanoparticles inks are categorized into four main categories: single element metallic nanoparticles inks, alloy metallic nanoparticles inks, metallic oxides nanoparticles inks and core-shell bimetallic nanoparticles inks. On the one hand, single element metallic nanoparticles inks are the most commonly used and widely available metallic nanoparticles inks in the market. Silver nanoparticles inks and gold nanoparticles inks are one of the few single element metallic nanoparticles inks used in the 3D printing of electronics due to their superb electrical conductivity and oxidation stability. On the other hand, the other three types of inks are less commonly used as they are not widely available in the market. However, some of these types of inks possess very interesting properties and have the potential to revolutionize 3D printing of electronics. The next section will discuss each type of metallic nanoparticles in greater detail.

3. Single Element Metallic Nanoparticles Inks

3.1. Silver Nanoparticles Inks

Silver has been used as money for centuries, and its silvery lustre makes it a perfect metal candidate for making jewellery. Silver is commonly known as a precious metal associated with hefty price tags. Apart from silver's monetary values and ornamental functions, silver is a unique metal that is very pertinent in many industrial applications: electrical, optical and biological areas. Silver is extensively used in electronics as excellent electrical conductors, and its unique optical properties also extended its uses in photovoltaic applications such as solar cells fabrications. Silver has remarkable antibacterial properties and is also being used in medical applications. Among all metals, silver has the highest thermal and electrical conductivity [21]. It has a thermal conductivity of 427 W/m·K [25] at 298.15 K, an electrical conductivity of 62.9×10^6 S/m [25] at 293.15 K and a melting temperature of 1234.93 K [26]. Despite these advantages, silver is prone to tarnish when exposed to hydrogen sulfide in air.

Silver nanoparticles inks are still extensively used for 3D printed electronics applications [4] due to their superior electrical conductivity and oxidation stability, despite their high costs [21, 27-28].

These conductive silver nanoparticles inks are available commercially in the market. Some of the major silver nanoparticles inks manufacturers [29] include Advanced Nano Products Co. Ltd. [30], Applied Nanotech Inc. [31], Clariant International Ltd [32-33], Creative Materials [34-35], Dupont [36], Harima Chemical Co. [37], Nova-Centrix [38-40], Paru [41], PV Nanocell [42], Sigma-Aldrich Corporation [43], Sun Chemical Corporation [44-45], UT Dots Inc. [46] and Xerox [47].

Most silver nanoparticles inks can be deposited onto substrates by inkjet and aerosol jet printers, and they typically require sintering temperatures ranging from 100 °C to 300 °C through thermal sintering in the ambient environment [48-49]. Silver nanoparticles inks can be used in many applications such as the fabrications of conductive patterns, strain gauges [5, 50], patch antennas [51], 3D antennas [52], radio-frequency identification (RFID) tags [53] and many more. However, the hefty price tags of silver nanoparticles inks result in difficulties achieving cost-effectiveness in industrial applications [28]. Therefore, one of the primary motivations is to find other alternatives which have lower costs and exhibit suitable electrical properties.

Silver nanoparticles also display unique optical, plasmonic and anti-bacterial properties [21, 54]. It is interesting to note that the optical, electrical and chemical properties of silver nanoparticles are also tunable, by merely modifying the particles' shapes, sizes, structures and other parameters [55]. The optical properties of the spherical silver nanoparticles are directly dependent on their diameters. The extinction spectra of spherical silver nanoparticles with diameters ranging from 10 nm to 100 nm shows that the spherical silver nanoparticles with diameters lesser than 50 nm have peak absorptance of light wavelengths at around 400 nm [56]. As the diameters of the spherical silver nanoparticles increase, the peaks broaden and shift towards longer wavelengths. These nanoparticles absorb the light at high efficiencies when they are exposed to the wavelengths of the light that corresponds to their peaks. Thus, these unique properties of silver can be further exploited to improve the qualities of the end-products of many printed electronics applications and as well as considering other interesting sintering techniques (such as intense pulse light (IPL) sintering [57-58] and ultraviolet (UV) sintering [59]) to sinter the deposited silver nanoparticles inks. Apart from the commonly-used spherical silver nanoparticles, nanosilver with unique geometries such as nanowires and nanoplates are also of great interests to researchers. Silver nanowires also can display up to 90% optical transparency and can be used to fabricate transparent electrodes [21]. However, silver nanowires have high tendency to clog up the inkjet printers' nozzles due to their geometries [60-61]. Silver nanoplates can form very dense microstructures due to their flat geometries, and hence allowing the printed patterns to have very good electrical conductance [21, 62].

3.2. Copper Nanoparticles Inks

Copper is a ductile and malleable metal with excellent electrical and thermal conductivity, which is generally used in the manufacture of electronics and wirings. Copper has reduced electromigration effects as compared to noble metals [28] and is also significantly cheaper as compared to silver and gold [63]. Copper has an electrical conductivity of 58.8×10^6 S/m [25] at 293.15 K, a thermal conductivity of 397 W/m·K [25] at 298.15 K and a melting temperature of 1357.77 K [26]. Although copper is only 6% electrically less conductive than silver [64], it is almost 100 times cheaper than silver. Therefore, many research interests are looking into the usage of copper nanoparticles inks for 3D printed electronics applications in recent years. However, the propensity of copper nanoparticles oxidising in a non-inert environment [28, 64-67] prevents extensive usage of pure copper nanoparticles in 3D printed electronics applications. The sintering of pure copper nanoparticles in the ambient environment at high elevated temperatures also expedites the oxidation process [67].

As oxidation takes place, thin coats of copper oxides (CuO) are formed on copper nanoparticles surfaces to achieve thermodynamic stability [68]. These undesirable copper oxides are found to be significantly less conductive than pure copper nanoparticles [19], and they also have a substantially higher melting point ($T_{\mathrm{m,\,CuO}}$ = 1603.15 K) than bulk copper too [69]. The electrical resistivity of bulk copper and bulk CuO are 1.72 μΩ cm and 5.1×10^{7} μΩ cm respectively [70]. Hence, viable electrons paths among the copper nanoparticles are significantly reduced [68] and lead to a decrease in electrical conductivity [65]. Also, it is also technically challenging to form highly conductive patterns by thermally sintering copper nanoparticles with oxides shells in the ambient environment [63]. As compared to silver and gold, copper's relatively higher melting point requires higher input amount of energy to coalesce copper nanoparticles to form conductive patterns [66]. Consequently, solving these challenges will unleash great potentials for fabricating cheaper electronics and enjoying significant cost savings, as compared to using silver or gold nanoparticles inks.

Nevertheless, there is literature indicating successful sintering of pure copper nanoparticles. Park et al. [28] formulated conductive copper nanoink for inkjet printing on glass substrates, thermally sintered the printed patterns in a vacuum chamber under 325 ° C for an hour and successfully achieved sheet resistivity of 17.2 μΩ cm. Despite the successful fabrication of conductive copper patterns, this complex and time-consuming method required a vacuum chamber [67] and involved sintering at high temperatures that might not be feasible for use with temperature-sensitive substrates [58, 63]. Apart from sintering the copper nanoparticles inks in a vacuum chamber, the copper nanoparticles inks can also be thermally sintered in inert gas (for instance, argon gas [70] and nitrogen gas [71]) or reducing gas conditions. There is difficulty in translating this time-consuming thermal sintering method into the electronic industry, which requires high-speed fabrications for mass production in the ambient environment. Literature also has shown that intense pulsed light (IPL) sintering [72-75] and laser sintering [72, 76] techniques are able to sinter copper nanoparticles inks at high speed in ambient conditions with low complexity, while maintaining high electrical conductance within the printed patterns. Niittynen et al. [72] sintered copper nanoparticles inks with both lPL and laser sintering techniques. He demonstrated that both techniques could achieve an electrical conductivity of more than 20% of bulk copper. However, the sintered copper nanoparticles inks are still vulnerable to oxidation and applications of protective layers are needed to prevent further oxidation and extend product life-time [77].

Copper nanoparticles inks are also commercially available via chemicals manufacturers like Applied Nanotech Inc. [78], and Nanoshel [79]. For instance, aerosol jet printable copper nanoparticles ink from Applied Nanotech Inc. (ANI Cu-OC70) [78] has particle size ranging from 20 – 100 nm, and it can achieve electrical resistivity between 20 – 50 μΩ·cm. ANI Cu-OC70 can be either be thermal sintered at above 350 °C in a reducing atmosphere (hydrogen, H_2 = 4% in nitrogen, N_2) for 20 minutes or sintered through the intense pulse light (IPL) sintering technique. Some applications with copper nanoparticles inks include conductive tracks, electrodes [75] and RFID tags [80].

3.3. Gold Nanoparticles Inks

Gold is a precious metal that has always captivated mankind for thousands of years for its highly lustrous appearances. Over the centuries, gold is also used as a common form of currency and in ornamental jewellery. Gold is a highly ductile and malleable noble metal, which can resist corrosion and oxidation and does not discolour or tarnish. Gold has a thermal conductivity of 314 W/m·K [25] at 298.15 K, an electrical conductivity of 41.0×10^6 S/m [25] at 293.15 K and a melting temperature of 1337.33 K [26]. Gold is also commonly used in the electronics industries for gold plating on other metallic conductors like copper and silver, to help prevent corrosions. However, it has an exorbitant price tag as compared to other metals.

The first usage of gold nanoparticles could be traced back from the 4th century A.D. of the late Roman empire and was demonstrated in the famous 'Lycurgus' Cup. Romans artisans embedded gold and silver nanoparticles into molten glass, creating a cup that was able to give different colours depending on the directions of the incident light [81]. In 1857, Michael Faraday demonstrated that by merely changing the particles size, it could alter the colour of the gold colloidal solutions [81-82]. Thus, implying that gold nanoparticles can have tunable optical properties. To date, gold nanoparticles are generally used in electronics, medical, biological, chemical and engineering applications areas. In electronics, gold nanoparticles are used for the fabrications of conductive tracks [83]. In the medical field, gold nanoparticles are adopted to be used as mechanisms for therapeutic agent delivery [84], diagnostics applications [85] and photodynamic therapy [83, 86]. Gold nanoparticles are even being exploited to be used as chemical sensors and optical probes in transmission electrons microscopy (TEM) [87].

Literature also indicated strong interests in using gold nanoparticles inks for printed electronics applications due to its good thermal stability, oxidation stability and electrical conductivity [66, 88]. However, gold nanoparticles inks are too expensive for cost-effective large-scale fabrications of cheap printed electronics. Nonetheless, gold nanoparticles inks are highly preferred in certain applications due to their unique properties. Tunable optical and electrical properties made gold nanoparticles to be one of the most unique metallic nanoparticles. These properties can be finetuned to each specific application needs, by directly modifying its parameters: compositions, sizes, shapes, structures and surface chemistry [54, 89-90]. Gold nanoparticles also exhibit interesting phenomenon like surface plasmon resonance (SPR) effects [91], which can be further exploited for the sintering process through photonic means (for instance, IPL and laser sintering techniques). Gold is highly resistant to oxidation, and it is very suitable for use in applications which require the electrodes to be resistant to oxidation due to external stimuli (for instance, electrochemical sensors [92] and wearable devices which may experience high salt concentration from sweat [88]). For printed organic thin film transistors (OTFTs) applications, OTFTs require very tight tolerances on their electrical resistances as slight variations may cause significant declines in their electrical performances [88]. Therefore, operationally stable gold nanoparticles inks are preferred for fabricating source and drain electrodes in OTFTs. Wu et al. [93] demonstrated the use of gold nanoparticles inks for the fabrication of highly conductive electrodes for organic thin film transistors (OTFTs) as most p-type organic semiconductors could form superb ohmic contacts with gold. Due to gold's good electrical conductivity, gold nanoparticles inks can also be used to fabricate conductive patterns, electrodes arrays [92, 94], microelectrode arrays (MEAs) for biosensing applications [88] and interdigitated electrodes (IDEs) [88]. It is also interesting to look into other shapes of gold nanoparticles other than spherical particles for printed electronics applications, such as gold nanorods [95] and nanowires [96]. For instance, mechanically flexible ultrathin gold nanowires have high optical transparency and are highly resistant to oxidation and corrosion. They can be utilised to fabricate flexible transparent electrodes [97] and flexible wearable sensors [96]. However, gold nanoparticles inks typically require sintering temperatures of more than 190 °C, which can limit the use of temperature-sensitive substrates such as polyethylene terephthalate (PET) for printed electronics applications [88]. Gold nanoparticles inks are commercially available via chemicals manufacturer like UT Dots Inc [98].

3.4. Aluminium Nanoparticles Inks

Due to its low cost and good mechanical properties, aluminium is used extensively in many industries such as aerospace, automobiles and even food and beverages industries. Aluminium is lightweight and highly corrosion resistant due to the thin films of protective aluminium oxides formed on its surfaces. Aluminium has an electrical conductivity of 35.5×10^6 S/m [25] at 293.15 K, a thermal conductivity of 238 W/m·K [25] at 298.15 K and a melting temperature at 933.47 K [26]. Its electrical conductivity is approximately 0.6 times of copper's electrical conductivity [99]. With the low material costs and compatible electrical properties of aluminium, aluminium nanoparticles inks are also synthesised for printed electronics applications. Conductive aluminium nanoparticles inks (Al-IS1000 [100] and Al-PS1000 [101]) are commercially available from Applied Nanotech Inc. From the manufacturer's datasheet [100], Al-IS1000 is an aerosol jet printable aluminium nanoparticles ink which has a viscosity ranging from 80 – 120 cP. Al-IS1000 is formulated for fabricating conductive patterns on silicon substrates, particularly for photovoltaic applications, as the ink has low contact resistivity on silicon substrates. In addition, Al-IS1000 is glass frit free, has passivation layer diffusion properties and can form highly uniform back surface field (BSF) layer. The printed aluminium film is said to be less than 10 mΩ/sq if sintered at the recommended temperatures ranging from 700° C to 900° C for less than a minute in air [100]. However, the high sintering temperatures restrict the usages of many temperature-sensitive substrates and may only allow substrates like glass or silicon to be used. This is also one of the main reasons why aluminium nanoparticles inks are not broadly adopted for 3D printed electronics applications. To date, there is few research in literature stating the usage of conductive aluminium nanoparticles inks for printed electronics applications. Khorramdel et al. [102] and Platt el al. [103] both discussed the use of aerosol jet printable aluminium nanoparticles inks, Al-IS1000, for fabrication of conductive tracks on silicon substrates.

3.5. Cobalt Nanoparticles Inks

Cobalt is ferromagnetic, corrosion resistant and commonly used for alloying. Cobalt also has good thermal and electrical conductivity, with a thermal conductivity of 100.0 W/m·K and electrical conductivity of 17.0×10^6 S/m [26]. Cobalt's melting temperature is at 1768 K[26]. Cobalt's high permeability and permittivity properties generate much research interests to formulate conductive cobalt nanoparticles inks for printed electronics applications, specifically to applications which require interactions with electromagnetic waves and high frequencies. However, cobalt nanoparticles inks for printed electronics applications are not available commercially. Nevertheless, some researchers formulate printable cobalt nanoparticles inks in-house with commercially available cobalt nanoparticles. Nelo et al. [104-105] formulated cobalt nanoparticles inks for screen printing applications and the printed patterns only required 10 minutes of sintering at 110 °C. Their cobalt nanoparticles inks had relative permeability and magnetic loss tangent values ranging between 1.5 – 3 and 0.01 – 0.06 respectively in the 45MHz – 10 GHz band [104]. These cobalt nanoparticles inks allow more explorations of printed electronics applications such as fabrications of radio frequency absorbers, antennas, magnetic sensors, filters, resonators and phase shifters [104-106].

3.6. Nickel Nanoparticles Inks

Similar to cobalt, nickel is a strong and corrosion resistant metal that able to withstand high temperatures. Nickel is the fifth most abundant metal on earth that commonly plays supporting roles for strengthening and stabilising other metals. It is commonly used in the plating industries, and it is also used for manufacturing superalloys and coins. Nickel metal is also ferromagnetic and commonly used for manufacturing permanent magnets. Nickel has good thermal and electrical conductivity, with a thermal conductivity of 91.0 W/m·K [26] and electrical conductivity of 14.3×10^6 S/m [26]. Nickel's melting temperature is at 1728 K [26].

Nickel's good electrical conductivity and ferromagnetic properties generate new interests in formulating printable nickel nanoparticles inks for printed electronics applications. Tseng et al. [107] studied the rheological and suspension structures properties of dispersed nickel nanoparticles in α-terpineol solvents and expected the printed patterns of these nickel nanoparticles inks to be conductive if printed on non-conductive substrates. Park et al. [108] formulated their nickel nanoparticles inks, and they sintered the printed patterns using the flashlight sintering technique. They were able to achieve a sheet resistance of 0.347 Ω/sq, which was suitable for printed electronics applications. Nickel is highly corrosion resistant and can be used as a barrier against oxidation [26]. Therefore, nickel nanoparticles inks can be used as a passivation layer to protect the underlying printed patterns (e.g. printed with copper and silver nanoparticles inks) from oxidation and corrosion for long-term operation. This can be done by merely depositing nickel nanoparticles inks over the underlying printed patterns. Furthermore, nickel nanoparticles inks are a lot cheaper than silver, gold and platinum inks. However, nickel nanoparticles inks tend to agglomerate and clog the inkjet printheads easily. Nickel nanoparticles inks are also commercially available in the markets, but only from a few vendors, such as Applied Nanotech Inc. [109-110] and Nano Dimension [111]. Applied Nanotech Inc.'s nickel nanoparticles inks: Ni-IJ70-30 [109] and Ni-OC70 [110], are inkjet and aerosol jet printable respectively. Their nickel nanoparticles inks can either be sintered through thermal sintering at temperatures more than 350°C in a reducing environment (H_2 = 4% in N_2) for 20 minutes or IPL sintering with the use of a xenon arc-discharge system. The sintered nickel nanoparticles inks can obtain an electrical resistivity ranging from 20 – 50 μΩ·cm, which are applicable for fabricating conductive tracks and patterns. Pasquarelli et al. [112] also reported depositing nickel nanoparticles inks onto glass and zinc oxide with an inkjet printer to fabricate electrode contacts for solar cell applications. Possible applications with nickel nanoparticles inks may include temperature sensors, inductors and microheaters.

3.7. Palladium Nanoparticles Inks

Palladium, a precious metal with silvery appearance, is commonly used in ornamental jewellery, dentistry for dental fillings and catalytic converters for automobiles. Palladium is also used in the fabrications of ceramic capacitors in the electronics industry. Chemical vapour deposition (CVD), sputtering and electroplating are some of the conventional methods to fabricate palladium structures [113]. Similar to all precious metals, palladium also has an expensive price tag. Palladium has a melting point of 1828 K, a thermal conductivity of 72 W/m·K and electrical conductivity of 9.5×10^6 S/m [26].

Due to palladium's unique reactions with hydrogen gas and its electrocatalytic properties [114], researchers are interested in synthesising palladium nanoparticles ink for printed electronics applications, particularly in fabricating electrochemical sensors. However, inkjet and aerosol jet printable palladium nanoparticles inks are not widely available in the market. Chemical manufacturers like SunChemical [115] and American Elements [116] do sell screen printable palladium inks for printed electronics applications. Liu et al. [113] filed a patent on the formulation of palladium nanoparticles inks. Their formulated palladium nanoparticles inks require a sintering temperature ranging between 180 °C – 250 °C for the coalescence of the palladium nanoparticles. Tseng et al. also looked into the deposition of palladium nanoparticles ink onto PET substrates through inkjet printing. The as-deposited palladium nanoparticles facilitate as a catalyst for electroless nickel plating at low operating temperatures [117]. Literature [118-120] also showed that palladium nanoparticles inks were commonly used as a catalyst for electroless plating of copper on polymer substrates for fabricating flexible electronics. Qin et al. [114] demonstrated the fabrication of pH sensors with their synthesized palladium precursor ink and thus implying that palladium nanoparticles inks also have the potential to be used for fabricating pH sensors. Possible applications with palladium nanoparticles inks may include electrode materials for fuel cells, pH sensors, conductive tracks, thin film transistors, gas sensors, biosensors and electrochemical sensors and monitoring systems [113-114].

3.8. Platinum Nanoparticles Inks

Platinum, one of the rarest noble metals, is corrosion resistant, malleable, ductile, highly dense and does not tarnish. Platinum metal is extensively used in engineering industries for applications like catalytic converters and electrical contacts. Due to its properties and rarity, platinum is also classified as a precious metal. It is commonly used for ornamental jewellery and is highly sought after for bullion investments. Therefore, hefty price tags are always tagged with platinum metal too. Platinum has a thermal conductivity of 71.6 W/m·K at 298.15 K, electrical conductivity of 9.1×10^6 S/m [25] at 293.15 K and melting temperature at 2041.4 K [26]. Due to platinum's unique properties, there are much research interests to explore printable platinum nanoparticles inks for printed electronics applications. Although platinum nanoparticles inks are not as accessible as silver nanoparticles inks or gold nanoparticles inks, manufacturers such as DuPont [121], Fraunhofer IKTS [122], LCC AkKoLab [122] and UT Dots Inc [98] do also manufacture these platinum nanoparticles inks. Most platinum nanoparticles inks can be deposited through aerosol jet or inkjet printing technologies [98]. The platinum nanoparticles inks typically require sintering temperatures ranging from 200°C – 250°C through thermal sintering [98]. However, the sintering temperatures are too high for the sintering of the platinum nanoparticles inks on temperature-sensitive substrates. In literature, Kim et al. used platinum nanoparticles ink to fabricate dye-sensitised solar cell (DSSC) counter electrodes [123]. Dupont's conductive platinum ink, BQ321, can also be used for printing of high activity electrodes in polymer thick film (PTF) sensors and biosensors [121]. Platinum nanoparticles inks can also be utilised for fabricating microelectromechanical systems (MEMS) gas sensors on thin ceramic substrates [123] and as well as electrodes for fuel cells applications.

3.9. Tin Nanoparticles Inks

Since the bronze age, archaeological records have revealed that people from ancient civilisations started using bronze for the creation of harder and durable tools and weapons [26]. Bronze is an alloy that primarily consists of copper and tin, and hence the earliest usage of tin is probably during the bronze age. In today's world, tin is commonly used for the manufacture of tin cans for the canning of processed food, whereby steel cans are plated with a thin layer of tin. Tin is corrosion resistant, and therefore tin is usually used to plate other types of metals that are prone to corrosions [26]. Tin has a melting point of 505.08 K, a thermal conductivity of 67 W/m·K and electrical conductivity of 8.7×10^6 S/m [26].

Although tin has a much poorer electrical conductivity as compared to silver and gold, it has significantly lower melting temperature and costs. Therefore, tin nanoparticles are ideal candidates for formulating low-cost conductive inks that are used for printed electronics applications. However, there are no commercially available tin nanoparticles inks. Jo et al. [124] synthesised tin nanoparticles inks for printed electronics applications and their conductive patterns were able to achieve an electrical resistivity of 64.27 μΩ·cm (six times higher than bulk tin) after 60 minutes of thermal sintering at 250 °C. The electrical resistivity of the printed patterns remained unchanged after 1 month and demonstrated the reliability of tin nanoparticles inks. Although the electrical resistivity of the tin nanoparticles inks are relatively higher than silver and gold nanoparticles inks, there are still potentials in using tin nanoparticles inks for printed electronics applications considering tin's price and melting temperature.

4. Alloy Metallic Nanoparticles Inks

4.1. Copper-Nickel Alloy Nanoparticles Inks

Copper-nickel alloy nanoparticles inks are receiving many interests recently for printed electronics applications due to their unique electrical properties. The nickel content in copper-nickel alloys helps to enhance the tensile strength, hot strength and proof strength, but it also results in a decrease in the electrical and thermal conductivity [125]. Copper-nickel alloy nanoparticles inks are also commercially available in the markets, but only from a few vendors, such as Applied Nanotech Inc. [126]. Applied Nanotech Inc.'s copper-nickel alloy nanoparticles inks: CuNi-OC5050 [126] and CuNi-IJ5050 [127] are aerosol jet and inkjet printable respectively. Their copper-nickel alloy nanoparticles inks can either be sintered through thermal sintering at temperatures more than 350°C in a reducing environment (H_2 = 4% in N_2) for 20 minutes or IPL sintering with the use of a xenon arc-discharge system [126]. CuNi-OC5050 can achieve a sheet resistance of 200 – 300 mΩ/sq for 2μm thin film according to the manufacturer [126]. Applied Nanotech Inc. also does sell copper-nickel alloy nanoparticles inks (CuNi-IJ5545) with 55/45 CuNi alloy ratio, which is similar to Constantan [127]. Constantan comprises 55% copper and 45% nickel, and it is the most widely used material for fabrication of strain gauges conventionally. Constantan possesses desirable properties like good fatigue life, high strain sensitivity and relatively high elongation capability [128]. Hence, the copper-nickel alloy nanoparticles inks are favorable to be used in sensing applications such as strain gauges and thermocouples [126].

5. Metallic Oxides Nanoparticles Inks

Metallic oxides nanoparticles inks, apart from single element metallic nanoparticles inks, are also gaining popularity in 3D printed electronics applications over time due to their superior oxidation stability [129] and hence, allowing the sintering of printed patterns in an ambient environment without the need for vacuum or inert gas conditions. Some of the commonly used metallic oxides nanoparticles inks for various functional printed electronics applications include copper oxide, iron oxide, indium-tin oxide and zinc oxide nanoparticles inks.

5.1. Copper Oxide Nanoparticles Inks

Although copper is significantly cheaper than silver, copper oxidizes readily in air to form copper oxides which made formulation and sintering of copper nanoparticles ink expensive and complicated. Hence, reducible copper oxide nanoparticles inks are one of the solutions to copper's oxidation and cost problems. These copper oxide nanoparticles are significantly cheaper than pure copper nanoparticles and easier to manufacture without worrying about oxidation. Reducible copper oxide nanoparticles inks comprise copper oxides nanoparticles and a reducer agent. Deposited reducible copper oxide nanoparticles inks are typically sintered by photonic sintering techniques in ambient conditions, in which the copper oxides nanoparticles are converted back into copper through a chemical reduction process. However, protective coatings should also be applied to the sintered printed patterns to prevent any potential oxidations [77]. Copper oxide nanoparticles inks are also commercially available via chemicals manufacturers like Nanoshel [130] and NovaCentrix [131-132]. For instance, inkjet printable copper oxide nanoparticles ink from NovaCentrix (Metalon ICI–002HV) [131] has copper oxide nanoparticles with particle size ranging from 110 – 130 nm, and it can achieve electrical resistivity between 7.5 – 10.8 μΩ·cm (approximately 4.5x bulk resistivity of copper). However, Metalon ICI–002HV can only be sintered via IPL sintering technique. Paquet et al. [133] demonstrated the use of IPL sintering technique on inkjet printed copper nanoparticles ink (NovaCentrix Metalon ICI–002HV) and were able to achieve an electrical resistivity of 9 μΩ·cm.

5.2. Iron Oxide Nanoparticles Inks

Iron oxide nanoparticles inks recently gained many interests among researchers for their magnetic properties (high permeability and relatively high saturation magnetisation) [134] and are further explored as functional printable magnetic inks for printable electronics applications such as inductors and radio frequency devices [135]. Vaseem et al. [135] formulated an iron oxide nanoparticle-based magnetic ink for the fabrication of a low-cost tunable frequency printed patch antenna.

5.3. Indium Tin Oxide Nanoparticles Inks

Indium tin oxide (ITO) is generally used for fabricating transparent electrodes in liquid crystal display (LCD), organic light emission display (OLED) and touch panels due to its high optical transmittance, electrical conduction, chemical inertness and good substrate adhesion [136-139]. There is also much interest to look into the use of inkjet printers for fabricating ITO thin films, as inkjet printing potentially offers better cost-effectiveness, material savings, direct patterning and printing efficiency as compared to current conventional fabrication techniques (for example, chemical vapor deposition (CVD), magnetron sputtering, and spin coating). There were several research teams [136-140] demonstrating the use of self-formulated ITO nanoparticles inks for fabricating transparent electrodes through inkjet printing. Hwang et al. [140] inkjet-printed 580 nm thick transparent electrically conductive films with ITO nanoparticles inks, and achieved a sheet resistance of 517 Ω/sq and optical transmittance of 87% after sintering them at 400 °C. In addition, they also sandwiched silver lines grid in between two layers of ITO thin films to further reduce the sheet resistance to 3.4 Ω/sq, but the optical transmittance was mildly reduced to 82% for the exchange of better electrical conductivity. However, ITO nanoparticles inks require high sintering temperatures which are not suitable for cheap temperature sensitive substrates.

5.4. Zinc Oxide Nanoparticles Inks

There are strong motivations to look for alternative materials to replace ITO due to the scarcity and high costs of indium, although ITO is an ideal material for fabricating transparent electrodes. Furthermore, ITO nanoparticles inks also require high sintering temperatures for well-optimised properties. Doped zinc oxide (ZnO) is one of the potential materials to replace ITO due to its superior material properties and low costs. ZnO is an n-type semiconducting material that has a wide bandgap of 3.36 eV and high optical transmittance, making it a potential material for fabricating transparent electrodes [141-142]. In addition, ZnO is also suitable for optoelectronics, bio-imaging, sensing, electronics, photovoltaics and sensing applications due to their unique piezoelectrical, luminescent and pyroelectrical properties [142]. Zinc oxide nanoparticles inks are also commercially available via chemicals manufacturer like Genes'Ink [143]. For instance, the inkjet printable zinc oxide nanoparticles ink from Genes'Ink (Helios'Ink H-SZ01034 semiconductive ink [144]) can achieve electrical conductivity between 10^{-8} – 10^{-7} S/cm before illumination. This ink is compatible with most flexible substrates, ITO and silver nanowires layers.

6. Core-Shell Bimetallic Nanoparticles Inks

In recent years, there is much on-going research exploring core-shell bimetallic nanoparticles (BNPs) inks for low-cost and high-performance 3D printed electronics applications [64]. Core-shell

BNPs consist of a core and a shell of two different elements, in which one type of metal atoms encircle the inner core that is made up of another type of metal atoms. Noble metals with good oxidation stability are usually used as outer shells in core-shell BNPs to protect the inner core from oxidations. Examples of core-shell BNPs include silver-gold (Ag-Au) [64], gold-palladium (Au-Pd) [64], tin-silver (Sn-Ag) [145] and copper-silver (Cu-Ag) [146-147] core-shell BNPs.

Core-shell BNPs are attractive to researchers due to the presence of additional unique properties that are not found in monometallic nanoparticles. Core-shells BNPs not only can give a combination of material, electrical, catalytic, optical and photocatalytic properties from two different metals, but also new properties arising from the synergy between both metals [148-149]. Core-shell BNPs also allow the tuning of the nanoparticles properties, in which specific desired properties are amplified and undesired properties are minimised. These properties can be altered by merely modifying the core-to-shell ratio of the core-shell BNP or changing its constituting materials [150]. With core-shell BNPs, it is also possible to reduce the expensive noble metal load in nanoparticles by substituting the inner core with cheaper metal alternatives. Costly noble metals are coated over cheaper metal alternatives (such as copper) to reduce the usage of costly monometallic noble monometallic nanoparticles for additional cost savings while maintaining similar electrical conductivities, oxidation stabilities and particle size [150]. However, synthesising these core-shell BNPs may be complicated and time-consuming.

6.1. Cu-Ag Core-Shell Bimetallic Nanoparticles Inks

On the one hand, silver nanoparticles inks are too costly to be used for large-scale fabrications of printed electronics despite their superior electrical and material properties. Silver is approximately

100 times more expensive than copper [151], and silver is only 6% more electrically conductive than copper. On the other hand, copper has excellent electrical properties, but copper nanoparticles are highly susceptible to oxidation in ambient environment at elevated temperatures. These copper oxides are undesirable for printed electronics applications due to their poor electrical conductivity and the need for higher sintering temperatures. Thus, copper nanoparticles are not very ideal for printed electronics applications. To simultaneously solve both cost issue in silver and oxidation issue in copper, it is particularly attractive and appealing to look into Cu-Ag core-shell BNPs inks for 3D printed electronics applications. The copper core is coated with a thin layer of silver shell in Cu-Ag core-shell BNP to help solve this oxidation problem of copper nanoparticles. As silver has high oxidation resistance, the silver shell can protect the copper nanoparticles from oxidation. Hence, with Cu-Ag core-shells BNPs, it is possible to significantly reduce the silver load in nanoparticles to enjoy better cost savings while maintaining similar electrical conductivities [146]. There is literature [146-147, 152-155] demonstrating successful uses of Cu-Ag core-shell BNPs inks for 3D printed electronics applications. These Cu-Ag core-shell BNPs inks demonstrate good electromigration and oxidation resistance properties. As silver is coated on the surface of the copper core, the required sintering temperatures of Cu-Ag core-shell BNPs inks are lower as compared to copper nanoparticles inks [155]. Cu-Ag core-shell BNPs inks are expected to have lower costs than silver nanoparticles inks due to their reduced silver loadings. However, these Cu-Ag core-shell BNPs inks are not available for sale commercially. Pajor-Świerzy et al. [152] reported on the synthesis of a low-cost conductive Cu-Ag core-shell nanoparticles ink for printed electronics applications. The Cu-Ag core-shell BNPs were made up of 1 μm diameter copper core coated with 20 nm thick silver shells. Their ink was reported to achieve 16% bulk conductivity of copper after 15 minutes of thermal sintering at 250 °C. Grouchko et al. [153] also demonstrated that

2 nm of silver layer was only required to coat the 40 nm copper nanoparticles to prevent oxidation and maintain the essential electrical properties of copper. These findings showed that the silver load could be significantly reduced with the use of Cu-Ag core-shell BNPs, as only thin shells of silver were needed to coat over the copper nanoparticles. Lee et al. [146] also synthesised Cu-Ag core-shell BNPs inks for printed electronics applications. Their results indicated that the Cu-Ag core-shell BNPs have better electrical properties and oxidation stability as compared to copper nanoparticles under ambient conditions, achieving 12 μΩ·cm at 350 °C.

6.2. Cu-Ni Core-Shell Bimetallic Nanoparticles Inks

As silver is an expensive material, some researchers have been looking into other metals such as nickel to replace the silver shells in Cu-Ag core-shell bimetallic nanoparticles inks. Kim et al. [156] explored the use of highly oxidation-resistant copper-nickel (Cu-Ni) core-shell BNPs inks for fabricating printed flexible electrodes. They achieved a reasonably low sheet resistance of 1.3 Ω sq^{-1} under the intense pulse light sintering process. Their research also showed that the printed electrodes still maintained their electrical properties after 30 days under 85% relative humidity at 85 °C. Hence, Cu-Ni core-shell BNPs inks can be further explored as an alternative conductive ink to the expensive silver nanoparticles inks in the future for cost-effective printed electronics applications.

7. Comparison of Different Metallic Nanoparticles Inks used in 3D Printed Electronics

Table 1 presents and compares the advantages and disadvantages of different metallic nanoparticles inks used in 3D printed electronics. From the table, it can be deduced that silver nanoparticles inks are still most suitable for fabricating highly electrically conductive patterns due to their better electrical conductivity and oxidation stability, despite their high costs. Furthermore, silver nanoparticles inks are widely available in the commercial market, and extensive research has been done by various manufacturers in the past few years to formulate silver nanoparticles inks for optimised depositions through the aerosol jet and inkjet printers.

It is also evident that silver and copper have the best bulk electrical conductivity among all metals. Although they are highly suitable for formulating highly conductive metallic nanoparticles inks, their melting temperatures are considerably high also. It is interesting to note that tin has a low melting temperature of 232°C and low material cost. Hence, it can be explored as an option for formulating low-cost conductive inks that only require low sintering temperatures.

Table 1. Comparison of different metallic nanoparticles inks used in 3D printed electronics.

Types of Metallic Nanoparticles Ink		Bulk Properties	Advantages	Disadvantages	Applications	Applicable Sintering Techniques	Reported Thermal Sintering Temperatures	Reported Electrical Properties	Inks Manufacturers or References	Commercially Available Inks
Single Element Metallic Nanoparticles Inks	Silver Nanoparticles Inks	• Electrical Conductivity: 62.9 × 10^6 S/m [25] • Thermal Conductivity: 427 W/ m·K [25] • Melting point: 1234.93 K [26]	• Excellent electrical conductivity, thermal conductivity and oxidation stability [21, 27-28] • Unique optical, plasmonic and anti-bacterial properties [21, 54] • Tunable optical, electrical and chemical properties [55] • Exhibits surface plasmon resonance (SPR) effects • Can be used in ambient environment • Widely available in the market	• Expensive • Difficulties in achieving cost effectiveness in industrial applications • Prone to tarnish when exposed to hydrogen sulfide	• Conductive tracks and patterns • Strain gauges [5, 50] • Patch antennas [51] • 3D antennas [52] • RFID tags [53]	• Thermal sintering [48-49, 157] • Laser sintering [158] • Intense pulse light (IPL) sintering [57-58] • Infrared (IR) sintering [159] • Ultraviolet (UV) sintering [59] • Microwave sintering [160] • Plasma sintering [161] • Electrical sintering [162]	• Sintering temperatures ranging from 100 – 300 °C [31, 48-49, 157]	• 3 μΩ·cm after 10 minutes of thermal sintering at 200 °C (UTDAgTE) [157] • 8 μΩ·cm after 60 minutes of thermal sintering at 140 °C (UTDAgTE) [157] • 10-50 μΩ·cm after 30 minutes of thermal sintering at 150 °C (Ag-IJ10) [31] • 9.1 μΩ·cm after 60 minutes of thermal sintering at 200 °C (Metalon® JS-A221AE) [163]	• Advanced Nano Products Co. Ltd. [30] • Applied Nanotech, Inc. [31] • Clariant International Ltd [32-33] • Creative Materials [34-35] • Dupont [29, 36] • Harima Chemical Co. [37] • Nova-Centrix [29, 38-40] • Paru [29, 41] • PV Nanocell [29, 42] • Sigma-Aldrich Corporation [29, 43] • Sun Chemical Corporation [44-45] • UT Dots Inc [46] • Xerox [29, 47]	• UTDAgTE (20-60 wt % Ag loading), UT Dots Inc. [157] • UTDAgPA, UT Dots Inc. [98] • UTDAgIJ, UT Dots Inc. [98] • UTDAgX, UT Dots Inc. [98] • Ag-IJ10 Nanosilver Ink (30 – 50 wt% Ag loading), Applied Nanotech, Inc. [31] • Metalon® JS-A221AE (50 wt% Ag loading), Nova-Centrix [163]
	Copper Nanoparticles Inks	• Electrical Conductivity: 58.8 × 10^6 S/m [25]	• Excellent electrical and thermal conductivity [64] • Reduced electromigration effects • Cheaper than noble metals	• Easily oxidises in air, which in turn significantly reduces the electrical conductivity of the printed patterns. [28, 64-67] • High melting point	• Conductive tracks and patterns • Electrodes [75] • RFID tags [80]	• Thermal sintering [78] • Intense pulse light (IPL)	• Thermal sintered at above 350 °C in a reducing atmosphere (H_2	• 5 – 7 μΩ·cm (Cu-IJ70) [164] • 20 – 50 μΩ·cm (Cu-OC70) [78]	• Applied Nanotech, Inc. [78] • Nanoshel [79]	• Cu-IJ70 Nanocopper Ink (30 – 50 wt% Cu loading), Applied Nanotech, Inc. [164]

		- Thermal Conductivity: 397 W/ m·K [25] - Melting point: 1357.77 K [26]	- Exhibits surface plasmon resonance (SPR) effects			sintering [72-75, 78, 164] - Laser sintering [72, 76]	= 4% in N$_2$) for 20 minutes [78]	- Achieve an electrical conductivity of more than 20% of bulk copper [72]	- Niittynen et al. [72]	- Cu-OC70 Nanocopper Ink (50 wt% Cu loading), Applied Nanotech, Inc. [78] - NS6130-04-476 Copper Nanoparticles Ink, Nanoshel [79]
	Gold Nanoparticles Inks	- Electrical Conductivity: 41.0×10^6 S/m [25] - Thermal Conductivity: 314 W/ m·K [25] - Melting point: 1337.33 K [26]	- Excellent electrical conductivity, thermal conductivity and oxidation stability [66, 88] - Tarnish and corrosion resistant - Does not discolor - Tunable optical and electrical properties [54, 89-90] - Exhibits surface plasmon resonance (SPR) effects [91]	- Very expensive - High melting point	- Conductive tracks and patterns - OTFTs [93] - Electrochemical sensors and electrodes [92] - MEAs [88] - IDEs [88] - Electrodes arrays - Flexible transparent electrodes [97] - Flexible wearable sensors [96]	- Thermal sintering [88, 165]	- Typically require sintering temperatures of more than 190°C [88] - Sintering temperatures of 200 – 400 °C (UTDAuTE) [165]	- 2 -7 x of bulk gold resistivity depending on temperature and time (UTDAuTE) [165]	- UT Dots Inc. [98]	- UTDAuTE - Gold nanoink for aerosol-jet printing using Ultrasonic Atomizer (20-60 wt % Au loading), UT Dots Inc. [98, 165] - UTDAuIJ, UT Dots Inc. [98] - UTDAuX, UT Dots Inc. [98]
	Aluminium Nanoparticles Inks	- Electrical Conductivity: 35.5×10^6 S/m [25] - Thermal Conductivity: 238 W/ m·K [25] - Melting point: 933.47 K [26]	- Low cost - Very good electrical and thermal conductivity - Corrosion resistant - Low contact resistivity on silicon substrates [100] - Passivation layer diffusion properties [100] - Can form highly uniform Back Surface Field (BSF) layer [100]	- High sintering temperatures required for printed conductive patterns	- Conductive patterns on silicon substrates, particularly for photovoltaic applications	- Thermal sintering [100]	- Low-temperature sintering: 550°C [100] - High-temperature sintering: 700-900°C for <1 minute in air [100] .	- <10 mΩ/sq* [100] - 80 mΩ/sq at both 550 and 600°C for 4 mins [103] - 20 mΩ/sq at 800°C for 4 mins [103]	- Applied Nanotech, Inc. [100]	- Al-IS1000 Aluminium Ink (Applied Nanotech, Inc.) [100]
	Cobalt Nanoparticles Inks	- Electrical Conductivity: 17.0×10^6 S/m [26] - Thermal Conductivity: 100 W/ m·K [26] - Melting point: 1768 K [26]	- Have ferromagnetic properties - Good electrical and thermal conductivity - Corrosion resistant [26] - High permeability - Required low sintering temperatures	- Much lower electrical conductivity as compared to silver and gold	- Radiofrequency absorbers - Antennas - Magnetic sensors - Filters - Resonators - Phase shifters [104-106] - Antenna miniaturization[104-105] - Antenna substrates [104-105] - Magnetic sensors or sensing[104-105]	- Thermal sintering [104-105]	- Thermal sintering at 350°C for 10 minutes [104-105]	- Relative permeability and magnetic loss tangent values ranging between 1.5 – 3 and 0.01 – 0.06 respectively in the 45MHz – 10 GHz band [104]	- Nelo et al. [104-105]	- Not Commercially Available

	Nickel Nanoparticles Inks	• Electrical Conductivity: 14.3×10^6 S/m [26] • Thermal Conductivity: 91.0 W/ m·K [26] • Melting point: 1728 K [26]	• Have ferromagnetic properties • Good electrical and thermal conductivity • Corrosion resistant • Cheap • Passivation properties	• Much lower electrical conductivity as compared to silver and gold • Tend to clog inkjet printheads due to agglomeration [111]	• Passivation layer for underlying silver and copper conductive patterns • Electrode contacts for solar cell applications • Conductive tracks and patterns • Temperature sensors • Inductors • Microheaters	• Thermal Sinteirng [109-110] • Intense pulse light (IPL) sintering [108-110]	• Thermal sintering at temperatures more than 350°C in a reducing environment (H_2 = 4% in N_2) for 20 minutes [109-110]	• 20 – 50 μΩ·cm [109-110] • 76.34 μΩ·cm [108] • 0.347 Ω/sq [108]	• Applied Nanotech, Inc. [109-110] • Nano-dimensions [111] • Pasquarelli et al. [112] • Tseng et al. [107] • Park et al. [108]	• Ni-IJ70-30 (30% Ni loading), Applied Nanotech Inc.[109] • Ni-OC70 (60% Ni loading), Applied Nanotech Inc.) [110]
	Palladium Nanoparticles Inks	• Electrical Conductivity: 9.5×10^6 S/m [26] • Thermal Conductivity: 72 W/ m·K [26] • Melting point: 1828 K [26]	• Good electrical and thermal conductivity • Unique reactions with hydrogen gas [114] • Have electrocatalytic properties • Unique material's properties for electrochemical sensors applications [113]	• Very expensive • Much lower electrical conductivity as compared to silver and gold • High melting point	• Electrode materials for fuel cells • pH sensors • Conductive tracks • Thin film transistors • Gas sensors • Biosensors • Electrochemical sensors • Monitoring systems [113-114]	• Thermal sintering [113, 115]	• Thermal sintering temperature ranging between 180°C – 250 °C [113] • Above 850 °C for at least 10 minutes (C2031105P2 - Screen Printable Palladium Ink Paste) [115]	• Not available	• Gwent Group, SunChemical [115] • American Elements [116] • Liu et al. [113] • Qin et al. [114]	• C2031105P2 - Screen Printable Palladium Ink Paste (67 - 68% Pd loading at 750°C), Gwent Group, SunChemical [115] • PD-M-01-INK – Screen Printable Palladium Ink, American Elements [116]
	Platinum Nanoparticles Inks	• Electrical Conductivity: 9.1×10^6 S/m [25] • Thermal Conductivity: 71.6 W/ m·K [26] • Melting point: 2041.4 K [26]	• Corrosion resistant • Does not tarnish • Good electrical and thermal conductivity • Unique material's properties for sensors, solar cells and biosensors applications • Inert • Good oxidation stability • Inkjet and aerosol jet printable [98]	• Very expensive • Very high melting temperature • Much lower electrical conductivity as compared to silver and gold	• Dye-sensitized solar cell (DSSC) counter electrodes • High activity electrodes in polymer thick film (PTF) sensors and biosensors • Ceramic MEMS gas sensors • Electrodes	• Thermal sintering [121, 166]	• Thermal sintering at 130°C for 5 - 10 minutes (DuPont BQ321) [121] • Thermal sintering at 300 °C for 30 minutes (UTDPtTE) [166]	• Sheet resistivity: 5 – 10 Ω/sq/mil (DuPont BQ321) [121] • Approx. 40 μΩ·cm at 300 °C for 30 minutes (UTDPtTE) [166]	• DuPont [121] • LCC AkKoLab [122] • UT Dots Inc [98] • Kim et al. [123]	• DuPont BQ321 - Screen Printable Platinum Ink, DuPont [121] • UTDPtTE (10-15% Pt loading), UT Dots Inc. [166] • UTDPtX, UT Dots Inc [98]
	Tin Nanoparticles Inks	• Electrical Conductivity: 8.7×10^6 S/m [26] • Thermal Conductivity: 67 W/ m·K [26] • Melting point: 505.08 K [26]	• Good electrical and thermal conductivity • Corrosion resistant • Low melting temperature • Cheaper cost [26]	• Much lower electrical conductivity as compared to silver and gold	• Conductive tracks and patterns [124]	• Thermal sintering [124]	• Thermal sintering at 250°C for 60 minutes [124]	• 64.27 μΩ·cm after 60 minutes of thermal sintering at 250°C (~ six times higher than bulk tin) [124]	• Jo et al. [124]	• Not Commercially Available

Alloy Metallic Nanoparticles Inks	Copper Nickel Alloy Nanoparticles Inks	- Not Available	- High tensile strength - Unique material's properties for strain gauges and thermocouples applications [126] - Inkjet and aerosol jet printable	- Much lower electrical conductivity as compared to silver and gold	- Conductive tracks and patterns [126] - Strain Gauges [126] - Thermocouples [126]	- Thermal sintering [126] - Intense pulse light (IPL) sintering [126]	- Thermal sintered at above 350 °C in a reducing atmosphere (H_2 = 4% in N_2) for 20 minutes [126]	- 200-300 mΩ/sq for 2μm film [126]	- Applied Nanotech, Inc. [126]	- CuNi-OC5050 (Available from 10-50 wt% loading), Applied Nanotech, Inc. [126] - CuNi-IJ5545, Applied Nanotech, Inc. [127] - CuNi-IJ5050, Applied Nanotech, Inc. [127]
Metal Oxides	Copper Oxide-Based Nanoparticles Inks	- Electrical Resistivity: 5.1×10^{7} μΩ·cm (CuO) [70] - Melting point: 1603.15 K (CuO) [69]	- Significantly cheaper than silver and copper nanoparticles inks - Good electrical conductivity - Able to sinter in ambient conditions	- Required photonic sintering techniques to sinter	- Conductive tracks and patterns	- Intense pulse light (IPL) sintering [131-132]	- Thermal sintering not applicable [131-132]	- 7.5 – 10.8 μΩ·cm (Metalon ICI–002HV) [131] - 9 μΩ·cm (Metalon ICI–002HV) [133]	- NovaCentrix [131-132] - Nanoshel [130] - Paquet et al. [133]	- Metalon ICI–002HV (16% CuO loading), Nova-Centrix [131] - Metalon ICI–003 (10% CuO loading), Nova-Centrix [132] - NS6130-10-1304, Nanoshel [130]

								- 13 – 15 μΩ·cm (Metalon ICI–003) [132] - 65-95 mΩ/sq (NS6130-10-1304) [130]		
	Iron Oxide Nanoparticles Inks	- Not Available	- Good magnetic properties [134]	- Low electrical conductivity [134]	- Inductors - Radio frequency devices - Patch antennas [135]	- Thermal drying [135]	- Drying at 80 °C for 30 min [135]	- Saturation magnetization of ≈12.4 under an applied field of 1 kOe [135]	- Vaseem et al. [135]	- Not Commercially Available
	Indium Tin Oxide Nanoparticles Inks	- Not Available	- High optical transmittance - Good electrical conduction - Chemical inertness - Good substrate adhesion [136-139]	- High sintering temperatures required for optimised transparency and electrical conductivity - Expensive [136-139]	- Transparent electrodes	- Thermal sintering [137-138] - Microwave Sintering [140]	- Thermal sintering at 600°C for 60 minutes [137]	- Sheet resistance of 2.19×10^{-3} Ω/sq and optical transmittance of 78.6%, after 60 minutes of thermal sintering at 600°C [137] - 50 – 300 Ω/sq [138] - Sheet resistance of 517 Ω/sq and optical transmittance of 87% after microwave sintering them at 400°C [140]	- Hong et al. [137] - Yu et al. [138] - Hwang et al. [140]	- Not Commercially Available
	Zinc Oxide Nanoparticles Inks	- Not Available	- Low costs - n-type semiconducting material - Wide bandgap of 3.36 eV - High optical transmittance	- Low electrical conductivity	- Transparent electrodes [141-142] - Optoelectronics, bio-imaging, sensing, electronics, photovoltaics and sensing applications [142]	- Thermal drying [141, 144]	- Thermally dried from 25 – 600°C for 15 – 60 minutes [141]	- 10^{-8} – 10^{-7} S/cm (H-SZ01034, Genes'Ink) [144]	- Genes'Ink [144] - Suganthi et al. [141]	- Helios' Ink H-SZ01034, Genes'Ink semi-conductive ink [144]

Core-Shell Bimetallic Nanoparticles Inks	Cu-Ag Core-Shell Bimetallic Nanoparticles Inks	• Not Available	• Better electrical properties and oxidation stability as compared to copper nanoparticles under ambient conditions • Potentially lower material costs as compared to silver nanoparticles inks due to reduced silver loading [146] • Lower initial sintering temperature as compare to copper nanoparticles inks [155]	• Not widely available in commercial markets • Synthesis of Cu-Ag core-shell BNPs inks may be complicated and time-consuming	• Conductive tracks and patterns	• Thermal sintering [146, 152-153]	• Thermal sintering at 250°C for 15 minutes [152]	• Achieve 16% bulk conductivity of copper after 15 minutes of thermal sintering at 250 °C [152] • Achieving 12 μΩ·cm at 350°C [146]	• Pajor-Świerzy et al. [152] • Grouchko et al. [153] • Lee et al. [146]	• Not Commercially Available
	Cu-Ni Core-Shell Bimetallic Nanoparticles Inks	• Not Available	• Highly oxidation-resistant • Lower material costs as compared to silver nanoparticles inks or Cu-Ag core-shell bimetallic nanoparticles inks	• Not widely available in commercial markets • Synthesis of Cu-Ni core-shell BNPs inks may be complicated and time-consuming	• Conductive tracks and patterns • Flexible electrodes [156]	• Intense pulse light (IPL) sintering [156]	• Thermal sintering not applicable [156]	• Achieved a reasonably low sheet resistance of 1.3 Ω /sq [156]	• Kim et al. [156]	• Not Commercially Available

*Dependent on sintering temperatures

8. Challenges

In spite of increasing demand for metallic nanoparticles inks, several challenges are required to be addressed to allow broader adoptions of metallic nanoparticles inks for 3D printed electronics applications.

8.1. High Costs of Silver Nanoparticles Inks

High costs of the commonly-used silver nanoparticles inks present a challenge for achieving cost effectiveness in industrial applications, but on-demand printing techniques such as inkjet and aerosol jet printing can help reduce material wastage to offset the high costs of silver nanoparticles inks. There is also much on-going research looking into finding cheaper alternatives to silver nanoparticles inks such as copper nanoparticles and copper oxide inks.

8.2. High Sintering Temperatures

Metallic nanoparticles inks typically require sintering temperatures above 100 °C to become electrically conductive in the thermal sintering process, and hence limiting the use of cheap temperature-sensitive substrates such as paper and common polymer foils for fabricating low-cost electronics. However, extensive research is being conducted in exploring alternative sintering techniques (for example, IPL and laser sintering) for low–temperature selective sintering to allow the use of temperature-sensitive substrates without severe damages or degradations.

8.3. Stretchability Issues

Metallic nanoparticles inks are not suitable for fabricating stretchable electronics due to their low stretchability characteristics, but they can be used for fabricating flexible electronics. On the one hand, soft substrates are highly stretchable and deformable. On the other hand, the sintered metallic nanoparticles inks are brittle and have low fracture strain [167]. Therefore, the primary technical challenge lies in the mismatch of the mechanical properties of the stretchable soft substrates and the sintered metallic nanoparticles inks where they adhere to each other. Hence, different types of inks such as conductive polymers are investigated to use for stretchable electronics applications.

8.4. Geometric Repeatability

3D printing of electronics with metallic nanoparticles inks inevitably face technical challenges in achieving void-free geometrically-uniform structures with smooth surface morphologies and well-defined edges. The coffee-ring effect is especially pertinent in inkjet-deposited metallic nanoparticles inks droplets, in which nanoparticles accumulate along the edges of the droplets [168]. Hence, 3D printed electrical components with consistent electrical properties and low tolerances may not be repeatable in mass productions, as morphology and geometry of the components play significant roles on their mechanical and electrical properties [4]. Therefore, more research efforts are required to solve these technical challenges to improve product reliability so that the electronic industry can accept printed electronics.

9. Outlooks and Potentials

The research trend is currently going towards fabricating printed stretchable electronics and sensors on soft substrates for wearable electronics and healthcare monitoring applications. However, metallic nanoparticles inks are not highly stretchable for these applications, and many researchers have been looking into other novel materials for fabricating stretchable electronics (for example graphene, graphene oxides and conductive polymers). Nevertheless, metallic nanoparticles inks still play an essential role in fabricating highly conductive patterns for circuitries on rigid and flexible substrates.

Apart from metallic nanoparticles inks, electrically conductive hybrid composite inks are also gaining significant attentions for various applications due to their ability to build conductive 3D structures [1]. Jo et al. [1] formulated a type of 3D printable electrically conductive hybrid composite ink, which comprised of carboxyl-terminated silver nanoparticles, silver flakes, amine-functionalized carbon nanotubes, and thermoplastic polystyrene–polyisoprene–polystyrene (SIS) triblock copolymer. Their material can achieve electrical conductivity of 22,939 S/cm and only require a sintering temperature of 80°C. This type of hybrid composite inks can allow researchers to explore the potentials of printing electrically conductive 3D structures, especially in antenna fabrications, to allow full optimizations of the available spaces.

Although there are many types of metallic nanoparticles inks in the market, they are not fully optimised and customized for printing with a specific printer on a particular substrate. Therefore, there is a need for greater co-operation efforts among inks and machines manufacturers to share their expertise. Nano Dimension [169] is one good example. They formulate and customize inks specifically for their inkjet printers (DragonFly 2020 Pro 3D Printer) for better optimisation and print quality [170]. To further expedite the progression of 3D printed electronics in revolutionizing industrial applications, research efforts should also focus on optimising the synergy between printing and sintering processes. These processes are interdependent with each other, and it is not possible to advance the 3D printed electronics technologies forward without concurrent research efforts (see **Figure 5.**). An ideal ink for electronics printing should possess these qualities: cost-effective, low sintering temperatures, favorable electrical and material properties, optimised for printers, and environmentally friendly. In addition, an ideal printer should also have high printing resolution, fast printing speed, on-demand non-contact printing to minimise contaminations, and ease of modifications and scalability of the printed patterns. At the same time, the sintering process should also possess high sintering speed, be compatible with the inks, gives excellent sintering properties and not damage the sintered patterns and substrates.

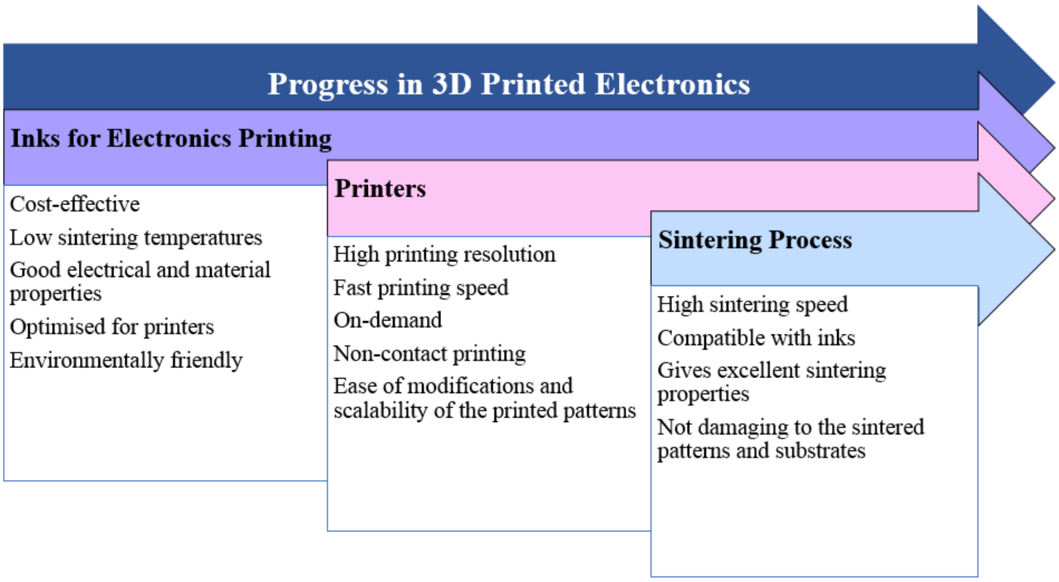

Figure 5. Ideal essential properties of inks, printers and sintering process for progress in 3D printed electronics.

10. Conclusion

3D printing of electronics shows great potentials in on-demand fabrications of customizable electronics, reducing the time taken for prototyping and exploring of highly innovative applications. This disruptive technology also aims to reduce wastage, time bottlenecks, costs and increase efficiencies as compared to the conventional ways of fabricating electronics. The emergence of various metallic nanoparticles inks in the market for 3D printed electronics has accentuated the growing 3D printed electronics sector in the additive manufacturing (AM) industry. Increasing adoptions of 3D printing by the electronics industry for fabricating electronics also increase the demand for various metallic nanoparticles inks with different material and electrical properties. This review paper presents a comprehensive overview of the different types of metallic nanoparticles inks used for 3D printing of electronics. Metallic nanoparticles inks are further categorized into single element metallic nanoparticles inks, alloy metallic nanoparticles inks, metallic oxides nanoparticles inks and core-shell bimetallic nanoparticles inks. The strengths and weaknesses of each type of ink are critically reviewed. The challenges and potentials of metallic nanoparticles inks in 3D printed electronics are also discussed. From the discussions above, core-shell bimetallic nanoparticles (BNPs) inks potentially seem to be the most cost-effective with materials and can give favorable electrical conductivity, good oxidation stability, and tailorable electrical and magnetic properties. Although core-shell BNPs inks may be complicated and time-consuming to synthesise with current state-of-the-art technologies, it is highly anticipated that future research, technological advancements and high-volume applications can significantly increase productivity and further drive down production costs with mass productions [77].

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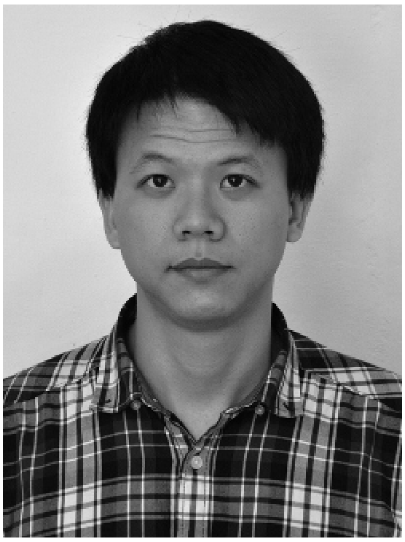

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