Review
Silver Nanoparticles for Conductive Inks: From Synthesis and Ink Formulation to Their Use in Printing Technologies

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Abstract: Currently, silver nanoparticles have attracted large interest in the photonics, electrics, analytical, and antimicrobial/biocidal fields due to their excellent optical, electrical, biological, and antibacterial properties. The versatility in generating different sizes, shapes, and surface morphologies results in a wide range of applications of silver nanoparticles in various industrial and health-related areas. In industrial applications, silver nanoparticles are used to produce conductive inks, which allows the construction of electronic devices on low-cost and flexible substrates by using various printing techniques. In order to achieve successful printed patterns, the necessary formulation and synthesis need to be engineered to fulfill the printing technique requirements. Additional sintering processes are typically further required to remove the added polymers, which are used to produce the desired adherence, viscosity, and reliable performance. This contribution presents a review of the synthesis of silver nanoparticles via different methods (chemical, physical and biological methods) and the application of silver nanoparticles under the electrical field. Formulation of silver inks and formation of conductive patterns by using different printing techniques (inkjet printing, screen printing and aerosol jet printing) are presented. Post-printing treatments are also discussed. A summary concerning outlooks and perspectives is presented at the end of this review.

Keywords: silver nanoparticles; synthesis; silver-based ink formulations; post-printing treatments

1. Introduction

Nanoparticles, defined as having one of the dimensions in the 1–100 nm range [1], show unique and considerably different physical, chemical, and biological properties, compared to bulk materials, due to their large portion of surface atoms and the reduced area to volume ratio. For many of them, enhanced reactivity and unique optical, electronic, and magnetic properties are observed [2].

Among various nanoparticle materials, silver nanoparticles (Ag NPs) have gained an enormous relevance due to their high electrical conductivity, low cost with respect to gold, relative resistance to oxidation, and biocidal properties [3,4]. Currently, many methods and approaches have been developed to synthesize Ag NPs, including chemical, physical, and biological routes, with the chemical methods being the most investigated and versatile. Physical methods, while producing nanoparticles with cleaner surfaces, are highly energy-intensive [5]. Biological synthesis has more advantages than the others. It
appears as a cost-effective and environmentally stable approach, but with higher limits in the reagents range and, therefore, on the morphological control. In this review, we try to bring out the possible ways that were successful in synthesizing Ag NPs for their use as fillers in the development of conductive inks.

In recent years, printed electronic devices have acquired great interest due to their low-cost high-volume manufacturing [6,7]. Compared with traditional production processes, such as evaporation, sputtering and chemical vapor deposition, which require complicated manufacturing processes and relatively high-cost equipment, printing technologies can bring competitive advantages in terms of product cost and flexibility, and performance [8,9]. There are a variety of printing technologies, including gravure printing, stereolithography, inkjet printing, screen printing, and aerosol jet printing. Each of these printing technologies has its advantages and disadvantages. The type of ink formulation and substrate plays an important role in determining the printing methods. In this review, three printing technologies (inkjet printing, screen printing and aerosol jet printing) are discussed.

Currently, silver-based inks are used in micro- or nano-electronic industries by printing technologies in a broad set of applications, such as sensors [10–12], solar cells [13,14], thin-film transistors [15], or supercapacitors [16,17]. The typical inks used for printing methods contain functional particles (e.g., Ag NPs), binders, solvents, and additives. Among these, functional particles, Ag NPs, are the main and the most important component. Ag NPs provide the electronic properties of the printed patterns. In general, the higher Ag NPs loading, the higher conductivity of printed patterns, and the conductive inks contain the solid loadings ranging from 20 to 80 wt.% [8]. In the case of Ag NPs inks, the size, shape, uniformity, and purity of Ag NPs determine the properties of the resulting products and should be considered to generate the final formulation and avoid undesired processing difficulties. The binder is often added into silver inks that helps in the homogeneous dispersion of Ag NPs into the ink. In addition, proper adhesion between printed patterns and substrates can be achieved by using the binder [18]. Solvents used as carriers for Ag NPs allow the ink to flow and impart favorable viscosity, surface tension, and homogeneity. A small quantity of additive is added to silver ink formulation and improves the property and performance of the ink. Generally, various additives, such as rheology modifiers, deformers, thickeners, and plasticizers, may be added to ink formulations, depending on the desired performance [19]. These components are coordinated together to the optimization of printing performance and quality of the printed patterns [20]. For instance, a not well-formulated ink could result in clogging of printer nozzles because of agglomeration or sedimentation of nanoparticles. The nanoparticles are then limited to a range of particle size and high colloidal stability to flow through the nozzles [8,21]. Alternatively, to solve the above issues, reactive silver inks have been proposed as a low-cost and promising solution for fabricating highly conductive silver tracks [22]. In the reactive inks, silver is a complex ion dissolved in an aqueous system, and low-temperature evaporation of complex ligands is sufficient for precipitating silver and reducing it to bulk silver for creating conductive patterns [23,24].

Physicochemical properties, such as viscosity, surface tension, evaporation rate and adhesion to substrates, are also very critically important to achieve high printing accuracy and resolution. The viscosity of conductive ink shows the behavior of its resistance to flow, and surface tension results from intermolecular forces that exist in liquids. For example, during the inkjet printing process, surface tension has an important effect on the formation of droplets. Higher surface tension values would result in earlier droplet breakup and faster drop velocity [25]. The viscosity and surface tension of conductive inks should be located in suitable ranges to meet the requirements of different printing technologies, which are presented later. The drying process removes moisture and/or solvents after the ink is transferred to the substrate without a significant increase in substrate temperature. The drying process imparts the proper adhesion of the ink to the substrate. There are many drying mechanisms, such as evaporation of water and/or solvent, penetration into the
printing substrate and air oxidation. Some inks combine two or more drying mechanisms during the drying process [26,27].

Polymers are relevant in ink formulations to promote good dispersion and avoid agglomeration, especially in metal-based nanoparticle inks. However, in order to produce highly conductive patterns, the dielectric polymer phase should be removed by sintering treatments [28].

Here, we will review the different methods to obtain Ag NPs, their use in the preparation of stable dispersion of inks depending on the printing method, and the post-processing of printed patterns for obtaining high electrical conductivity, as shown in Figure 1.

Figure 1. Preparation of silver inks, different printing techniques and sintering methods.

2. Synthesis of Ag NPs

The size, shape, uniformity, and stability, as well as the corresponding physical, chemical, and biological properties of nanoparticles, are strongly dependent on synthetic methods [2]. A variety of methodologies are employed to synthesize Ag NPs, generally classified in chemical, physical, and biological methods [29]. The different methods are described in the following sections.

2.1. Synthesis of Ag NPs by Chemical Methods

Chemical methods are the most extensively investigated route for developing Ag NPs. The chemical methods can be divided into four different categories: (1) chemical reduction, (2) electrochemical techniques, (3) pyrolysis, and (4) irradiation-assisted methods [30]. Among these methods, the chemical reduction of Ag\(^{+}\) species to Ag\(^0\) in solution using reducing agents is the most common and widely reported synthesis method of Ag NPs, usually no aggregation, high yield, and low preparation cost [29].

The formation mechanism of nanoparticles from solutions has been investigated, including nucleation and growth process [31]. The nucleation process requires more activation energy than the growth process, which is controlled by diffusion. The relative rates of nucleation and growth process are governed by varying reaction parameters, such as concentration of the reactants, the potency of the reducing agent, pH, or temperature. These parameters can be adjusted to fine-tune the nanoparticle synthesis achieving a single nucleation step and a steady growth process following a typical formation pattern of a LaMer model [32]. Figure 2 shows a plot of the LaMer model in which the monomer concentration is shown as a function of time [33]. In Stage I, the concentration of free monomers in solution increases sharply until a supersaturation (C\(_s\)) point is obtained. The
concentration of monomer is high enough to overcome the energy barrier for nucleation. In Stage II, burst nucleation leads to a significant decrease of the monomer concentration in solution, which reaches below the level of minimum supersaturation ($C_{\text{min}}$), where no additional nucleation occurs. In Stage III, growth continues without further nucleation to form nanocrystals. The separation of nucleation and growth process gives rise to identical growing times of all the nanoparticles, and therefore, uniformity in sizes. As an alternative to this synthesis, a very high uniformity can be achieved using seed-mediated-growth synthesis instead of seedless synthesis. Small nanoparticles, seeds, are added to a growing solution and act as the nucleus for continuous growth. The reduction kinetics, in this case, is maintained to be slow enough not to achieve saturation.

![Schematic illustration of the LaMer model for the generation of monomers, nucleation, and subsequent growth.](image)

The chemical reduction method usually involves three main components, i.e., metal precursors, reducing agents, and stabilizing/capping agents [34]. Different reducing agents, such as sodium citrate, ascorbate, sodium borohydride (NaBH$_4$), elemental hydrogen, polyol, Tollén’s reagent, N, N-dimethylformamide (DMF), or polyethylene glycol (PEG)-block copolymers, are used. The choice of reducing agents and solvents gives rise to a different type of synthesis and offers an enormous number of possibilities.

Among the various possibilities, Turkevich’s and Brust’s methods, using citrate and NaBH$_4$, are the most used ones and generate very different nanoparticles in terms of sizes and solvent solubility [35,36]. Together with the choice of metal salt, reduction agent, and experimental factors, the capping agent is the main factor in determining the size and physicochemical properties of the nanoparticles [37]. This capping provides colloidal stability to the nanoparticle, either electrostatically or sterically, and controls the nanoparticle growth due to interactions with different facets, generating different morphologies. Capping agents contain polymers, such as poly (N-vinyl-2-pyrrolidone) (PVP), PEG, polymethacrylic acid (PMAA) and polymethylmethacrylate (PMMA) or small molecules such as citrate, oleilamine, alkanethiols, etc. Different morphologies (nanospheres [38], nanorods [39], nanocubes [40], nanodisks [41], nanoprisms/nanotriangles [42], truncated octahedron [43], nanobipyramids [44], nanoshells [45,46], nanobars [47], nanorice [47], nanowires [48] and nanostars [49]) have been achieved by selecting capping agents and
controlling experimental parameters, as shown in Figure 3. The nanoparticle shape controls, among others, the plasmonic modes and offers the possibility to manipulate and respond to a light stimulus that goes from 400 nm to the middle infrared [50,51]. The morphology is also relevant in their catalytic activity and the creation of electrical paths in conductive composites and inks, where high aspect ratio morphologies are preferred.

![Figure 3. Ag NPs with different shapes.](image)

The synthesis parameters (reaction temperature, pH of precursors, reducers and stabilizing agents) are crucial factors in determining nanoparticles formation and nucleation process. Many different examples of Ag NPs synthesis were proposed. Wiley et al. [52] obtained Ag NPs with three different shapes (nanocubes, bipyramids, nanowires) using ethylene glycol as a reducing agent. In their experiment, PVP was added as a stabilizing agent and reducing agent, which also played a role in controlling the shape. In addition, during the reaction, the seeds with different crystal structures were selected via etching, which produced the different morphologies of Ag NPs. Xue et al. [53] reported pH-switchable silver nanoprism growth pathways. They found that excellent control over the growth of Ag nanoprisms can be achieved by adjusting solution pH. Jiang et al. [54] investigated the reaction temperature effect on the formation and growth of Ag NPs using two or three reducing agents, including citric acid, L-ascorbic acid, and NaBH₄. They found that there was a significant jump in the particle size at around 32 °C. The size of plates increases from 90 to 180 nm and from 25 to 48 nm for spheres. Yoo et al. [55] synthesized Ag NPs via hydrazine reduction of Tollen’s reagent. In their work, AgCl and ammonium hydroxide were mixed in the presence of PVP as a stabilizer, resulting in a clear solution without any precipitate. Then, hydrazine hydrate was injected to reduce [Ag(NH₃)₂]⁺ and obtain Ag NPs at room temperature. The size of the obtained Ag NPs was controlled by the concentration of PVP. The diameter of Ag NPs ranged from 68 to 119 nm. In addition, AgCl was retrieved from the electronic scrap and used as a metal precursor. Zhang et al. [56] prepared colloid Ag NPs by using hyperbranched poly(methylene bisacrylamide-aminoethyl piperazine) with terminal dimethylamine groups (HPAMAM-N(CH₃)₂) as the effective reducing and stabilizing agent. The Ag NPs were synthesized by mixing AgNO₃ aqueous solution with HPAMAM-N(CH₃)₂ aqueous solution at room temperature. A series of Ag NPs was prepared by adjusting the molar ratio of N/Ag in feed. Malassis et al. [57] used one-step aqueous synthesis of Ag NPs using ascorbic acid (vitamin C) as a reducing and stabilizing agent at the same time. Metal precursor, AgNO₃, is mixed with ascorbic acid
under a strong stirring at room temperature for 30 s. The authors showed that the size of Ag NPs obtained in the range of 20 to 175 nm was affected by the pH of either the metal salt solution or the ascorbic acid solution. Leng et al. [58] produced Ag NPs by the reduction of AgNO$_3$ using the ethylene glycol in the presence of PVP. They also studied the different reaction conditions (i.e., AgNO$_3$ concentration, AgNO$_3$ and PVP mass ratio and the reaction temperature) on the size and size distribution of Ag NPs. When the mass ratio of AgNO$_3$ to PVP was 2:1, Ag NPs with a narrow size distribution from 20 nm to 30 nm were obtained. Sakthivel et al. [59] prepared isonicotinic acid hydrazide capped Ag NPs by the wet chemical method and used NaBH$_4$ as a reducing agent. The synthesized Ag NPs distributed in two ranges and the average size of the particles is 35 and 536 nm. Isonicotinic acid hydrazide capped Ag NPs were applied to detect Hg$^{2+}$ in an aqueous medium in the nanomolar range. Sreelekha et al. [60] synthesized Ag NPs using trisodium citrate as reducing and stabilizing agents via the chemical method. As described in their work, Ag NPs were prepared by mixing the trisodium citrate solution and AgNO$_3$ solution at a temperature of about 80 °C, followed by separation of the NPs by centrifugation and drying to collect. Kamarudin et al. [61] fabricated two types of Ag NPs by varying the amount of precursor and synthesis temperature. The higher reaction temperature, the higher reaction rate, which increased the process of ions to NPs conversion.

Waqas et al. [62], for instance, successfully fabricated Ag NPs with three different shapes (i.e., spherical, star and pyramidal) via a simple wet chemical approach. An aqueous solution containing silver nitrate (AgNO$_3$), sodium hydroxide, oleic acid, and ammonia was used in their synthesis. During the reaction, the concentration of sodium hydroxide and oleic acid played an essential role in controlling the size, shape, and homogeneity of Ag NPs. Among all the three shapes, Ag NPs with star shape showed the highest electrocatalytic sensing efficacy in terms of linear range, the limit of detection, and sensitivity, because of more exposed catalytic active sites and fast diffusion of ions between electrode-electrolyte interfaces.

Meanwhile, Makwana et al. [63] successfully prepared fluorescent Ag NPs by only applying a single chemical reagent calix [4] resorcinarene polyhydrazide (CPH) which acted as a reduction and dispersant agent. Bearing hydrazide group of CPH on its periphery acted as a reducing agent, and its web type of structure as a stabilizing agent for the formation of calix-protected Ag NPs. Their results showed a large quantity of well-dispersed spherical CPH-Ag NPs with an average diameter of 7 nm. In addition, CPH-Ag NPs were not only selective and sensitive fluorescent sensors for Fe$^{3+}$ from 0.1 to 10 µM but also exhibited reasonably good antimicrobial activity.

Bare Ag NPs are not stable and can rapidly undergo agglomeration due to their high reactivity. In most of the preparation routes, capping agents are required to control the particle size, from absorbing or attaching with each other surfaces and avoiding the aggregation of nanoparticles; coating is a way to produce electrostatic, steric or electrosteric interactions between particles and helps stabilize the nanoparticles [64,65]. Conversely, the capping agents often influence various properties of nanoparticles, including their shape and interactions with surrounding solvent, therefore, its dispersibility and size [37]. For instance, Ajitha et al. [37] investigated the influences of capping agents (PEG, ethylenediaminetetraacetic acid (EDTA), PVP, and polyvinyl alcohol (PVA)) on Ag NPs using a simple chemical reduction method. As described in their research, the average particle size was the largest for the PEG-capped NPs with a diameter of 44 nm and the smallest for the PVA-capped ones with a diameter of 27 nm. In addition, the PVA-capped ones were observed to be the most stable and to display the highest antibacterial activity. Other examples for the synthesis of Ag NPs by chemical methods are given in Table 1.
Table 1. Synthesis of Ag NPs using chemical methods.

| Precursor      | Reducing Agent                  | Stabilizer Agent               | Size       | Structure/Shape        | Reference |
|----------------|---------------------------------|--------------------------------|------------|------------------------|----------|
| AgNO₃          | Gallic acid                     | Gallic acid                    | 89 nm      | Pseudospherical        | [66]     |
| AgNO₃          | NaBH₄                            | Tartrate, citrate and PVP      | 118 ± 18 nm| Tetrahedral            | [67]     |
| AgNO₃          | Hydrazine hydrate and sodium citrate | Sodium dodecyl sulfate          | 40–60 nm   | Spherical              | [68]     |
| AgNO₃          | Sodium citrate and tannic acid   | PVP                            | 10–200 nm  | Spherical              | [69]     |
| [Ag(NH₃)₂]⁺    | Ascorbic acid                   | Citrate                        | 40–300 nm  | Spherical              | [70]     |
| AgNO₃          | Hydrazine hydrate               | PVP                            | 300 nm     | Spherical              | [71]     |
| AgNO₃          | Hydrazine                       | Trisodium citrate              | 35 nm      | FCC and spherical      | [72]     |
| AgNO₃          | Sodium citrate                  | PEG                            | 50 ± 19 nm | Spherical, triangular and rod | [73]     |
| AgNO₃          | Dextran                         | Dextran                        | 15 nm      | Spherical              | [74]     |
| AgNO₃          | Ascorbic acid                   | Surfactin                      | 15–35 nm   | Spherical              | [75]     |
| AgNO₃          | NaBH₄                           | PVP                            | 1–75 nm    | FCC                    | [76]     |
| AgNO₃          | Plasma                          | Trisodium citrate              | 35–100 nm  | FCC and spherical      | [77]     |
| AgNO₃          | Sodium citrate                  | Sodium citrate                 | 20–60 nm   | Spherical              | [78]     |
| AgNO₃          | NaBH₄                           | Chitosan                       | 10–100 nm  | Spherical              | [79]     |
| AgNO₃          | NaBH₄                           | Sodium citrate                 | 60 nm      | Spherical              | [80]     |

2.2. Synthesis of Ag NPs by Physical Methods

Physical methods usually are fast, do not involve toxic chemicals, and form a relatively narrow distribution of the synthesized Ag NPs size. However, they present several disadvantages compared with chemical methods, such as less control on the nanoparticle morphologies, more significant difficulties for the scaling and the requirement of more sophisticated equipment during the synthesis process. There are several different physical methods to synthesize Ag NPs, such as evaporation-condensation [30], ball milling [81], arc discharge [82], laser ablation [83], and pulsed wire evaporation (PWE) [84]. Other examples for the synthesis of Ag NPs by physical methods are given in Table 2.

Table 2. Synthesis of Ag NPs using physical methods.

| Physical Method | Medium            | Size       | Structure/Shape      | Reference |
|-----------------|-------------------|------------|----------------------|----------|
| PWE             | Deionized Water   | <100 nm    | Spherical            | [84]     |
| Laser ablation  | Deionized Water   | 7 ± 3 nm   | Spherical            | [85]     |
| Arc Discharge   | Ethanol           | ≈20 nm     | Spherical            | [86]     |
| Laser ablation  | Deionized Water   | 15 nm      | Spherical            | [87]     |
| Laser ablation  | Deionized Water   | 21 nm      | FCC and semi-spherical | [88] |
| Laser ablation  | Methanol          | 26 ± 5 nm  | FCC and spherical    | [89]     |

The PWE method is known as a one-step synthetic technique to obtain nanoparticles through evaporation and condensation processes with high efficiency and high production rate [90], as shown in Figure 4. In the PWE method, electrical energy with a high voltage and high electric current is applied through a thin metallic wire, resulting in the heat and vaporization of the thin wire. Then, nanoparticles can be obtained by condensation through
a peripheral cooling gas or liquid. The product mass and size of nanoparticles can be controlled by changing the wire explosion number, voltage, and current [84].

![Schematic of PWE](image)

**Figure 4.** Schematic of PWE. In this technique, a high voltage is applied at the tip of the wire in a liquid solution, generating nanoparticles after evaporation and condensation.

Song et al. [91] investigated the effect of variation in voltage on the morphology and microstructure of Ag NPs synthesized by PWE. The average particle size increased by increasing the supply voltage without any impurity and oxide phase. The obtained Ag NPs exhibited an almost spherical shape and very small particle size (less than 30 nm). In addition, Chung et al. [92] fabricated silver/copper combined nanoparticles by one-step electrical explosion of metal wires. The silver/copper nano inks were printed on flexible polyimide (PI) and sintered via the flash white light process. The sintered films show a lower resistivity (4.06 µΩ·cm) and oxidation stability than the copper nanoparticle film.

### 2.3. **Synthesis of Ag NPs by Biological Methods**

The drawbacks with the chemical and physical methods of Ag NPs formation are that they are highly costly and involve the use of toxic and hazardous chemicals. They contain potential environmental and biological hazards [93]. Therefore, the development of the biological synthesis of Ag NPs is advancing as a critical branch of nanotechnology. The use of biological entities, such as plant extracts, bacteria, actinomycetes, fungi or enzymes, for the generation of nanoparticles could be alternative to chemical and physical methods [94]. The advancements of biological synthesis of nanoparticles over chemical and physical methods are cost-effective and easily scaled up for vast-scale synthesis of nanoparticles, while high temperature and harmful solvents and reagents are not required for biological synthesis [95]. Compared with traditional chemical synthesis, the disadvantages are the limitation of reagents that makes it difficult to control the size and, more specially, the morphology of the nanoparticles, and the higher amounts of impurities compared with physical methods.

Biological synthesis using plants and plant extracts appears a relatively straightforward and advantageous approach compared with microorganisms, such as bacteria and fungi, due to its single-step technique, economical protocol and non-pathogenicity [94]. Plants have been extensively applied for the green synthesis of Ag NPs because plant extracts can act as both reducing and stabilizing agents, decreasing the cost and simplifying the process. Various plant resources, such as bark, roots, stems, fruits, seeds, callus, peels,
leaves and flowers, have been explored by researchers till date to synthesize Ag NPs [95–97]. Additional steps such as extraction protocols or mixing extracts from different plants are also common in this type of synthesis.

Although the number of biological syntheses has grown rapidly in the last years, the synthesis protocols are still much less than in the case of chemical synthesis. Among the different examples, Anandan et al. [98], for instance, synthesized Ag NPs with distinct morphological characteristics using leaf exacts of Dodonaea viscosa as reducing agents. Five different polar and non-polar solvents (petroleum ether, methanol, acetone, acetonitrile and water) were used for the extraction of active ingredients from the leaves of Dodonaea viscosa, which attributed to obtaining nanoparticles with different properties. The high resolution-transmission electron microscopy (HR-TEM) analysis showed different nano sizes (15, 18, 12 and 20 nm) of the nanoparticles with different surface morphologies (worm-like, irregular flower, spherical and dendritic structures) using methanol, acetone, acetonitrile and water extracts, respectively.

A cost-effective and straightforward biosynthesis of Ag NPs was reported by Hassenin et al. from the purple heart plant [99]. The aqueous methanolic extract was prepared and employed to synthesize Ag NPs by the biological reduction method using an aqueous solution of AgNO₃. As described in their research, the effect of various independent factors (concentration of AgNO₃, temperature, and the volume of leaves extract solution) on particle size, polydispersity index of synthesized Ag NPs was optimized using a Box–Behnken design. The synthesized Ag NPs at optimum condition are spherical with a particle size of 98 nm, as observed by TEM.

Kalaiselvi et al. [100] revealed that the latex extract of Euphorbia tirucalli can produce Ag NPs against the root-knot nematode species, Meloidogyne incognita. The biosynthesized Ag NPs are spherical and cubic, with the particle size in the range of 20–30 nm. Tripathi et al. [101] synthesized Ag NPs utilizing in vitro grown leaf extract of medicinal plant Withania coagulans by reducing AgNO₃ solution. The results indicate that nanoparticles were spherical with an average size of 14 nm and had face-centered cubic (FCC) structure. In addition, prepared Ag NPs have antibacterial activity against both gram positive and negative bacteria. Ramesh et al. [102] reported that Ag NPs were synthesized using an aqueous leaf extract of Ficus hispida Linn. ft. The formation rate of Ag NPs depended on the concentration of AgNO₃, concentration of leaf extracts, temperature, pH and reaction time. The synthesized nanoparticles were spherical with an average particle size of 20 nm. Other plants and plant extracts used for the synthesis of Ag NPs are given in Table 3.

| Plants/Plants Extracts | Precursor | Reducing and Capping Agent | Size    | Structure/Shape                  | Reference |
|------------------------|-----------|----------------------------|---------|----------------------------------|-----------|
| Rheum palmatum         | AgNO₃     | Root extract               | 121 ± 2 nm | Hexagonal, spherical, and cubic | [103]     |
| Handelia trichophylla  | AgNO₃     | Shoot extract              | 20–50 nm | FCC and spherical                | [104]     |
| Turmeric powders       | AgNO₃     | Extract                    | 18 ± 0.5 nm | Spherical                        | [105]     |
| Bilberry and Red Currant Waste | AgNO₃ | Fruit extract | 25–65 nm | FCC                              | [106]     |
| Green tea extract      | AgNO₃     | Extract                    | 3.9 ± 1.6 nm | FCC and spherical                | [107]     |
| Reishi mushroom        | AgNO₃     | Extract                    | 15–22 nm | FCC and spherical                | [108]     |
| Alternanthera dentata  | AgNO₃     | Leaf extract               | 10–80 nm | FCC                              | [97]      |
Plant Extracts Precursor Reducing and Capping Agent Size Structure/Shape Reference

| Plants/Plants Extracts | Precursor | Reducing and Capping Agent | Size | Structure/Shape | Reference |
|------------------------|-----------|----------------------------|------|-----------------|-----------|
| Plants from Myrtaceae family | AgNO\(_3\) | Leaf extract | 5–55 nm | Spherical | [109] |
| Andrographis paniculata, Phyllanthus niruri, and Tinospora cordifolia | AgNO\(_3\) | Stem and leaf extract | 50–12 nm | Spherical | [110] |
| Stachytarpheta cayennensis | AgNO\(_3\) | Leaf extract | 13 nm | FCC, and spherical | [111] |
| Memecylon umbellatum Burn F | AgNO\(_3\) | Leaf extract | 7–23 nm | Spherical | [112] |
| Galega officinalis | AgNO\(_3\) | Leaf extract | 8–34 nm | FCC and spherical | [113] |
| Portulacaria afra | AgNO\(_3\) | Leaf extract | 27 ± 4 nm | Irregular spherical | [114] |
| Cleome viscosa L. | AgNO\(_3\) | Fruit extract | 20–50 nm | Spherical | [115] |
| Origanum vulgare L. | AgNO\(_3\) | Extract | 2–25 nm | FCC and spherical | [116] |

3. Silver Inks for Printing Techniques

The typical inks used for printing methods contain functional particles (e.g., Ag NPs), binders, solvents, and additives. The common strategy for the formulation of silver inks is to add silver nanomaterials into polymers dissolved in suitable solvents to fabricate conductive inks. In some cases, this polymer could be the same as the capping agent that stabilizes the nanoparticles. In order to protect Ag NPs in inks against aggregation and oxidation, organic stabilizers/polymer is required. Then, a high-temperature sintering step is often needed to remove organic stabilizers and improve the conductivity, taking into account that the temperature may affect the substrate. More recently, a novel kind of metallic inks has been developed, called reactive ink, as a low-cost and promising alternative for fabricating highly conductive silver tracks. Reactive silver ink is a pure solution-phase ink mainly composed of a silver salt precursor and complexing agent. After printing, conductive silver can be generated through a reduction reaction and/or by the decomposition of a metal-organic precursor [28].

It should be noted that different performance and properties of conductive inks should be used by different printing technologies depending on the type of printing technology being used and the desired printed performance. In this review, three printing technologies (inkjet printing, screen printing and aerosol jet printing) are discussed. The inkjet printer ejects droplets of inks as required through the printing nozzle on a variety of substrates, as shown in Figure 5. There are two different modes of how the droplet is jetted from the nozzle, i.e., continuous inkjet printing and drop-on-demand printing. The optimum viscosity and surface tension for inkjet printing should be in the range 0.001–0.1 Pa·s and 15–25 mN·m\(^{-1}\), respectively [28]. Screen printing is the process that transfers the inks pass through the patterned stencil with a squeegee, as shown in Figure 5. The optimum viscosity and surface tension for inkjet printing should be in the range 0.5–5 Pa·s and 0.001–0.1 Pa·s and 38–47 mN·m\(^{-1}\), respectively [28]. Aerosol jet printing is the process that involves forming an ink aerosol and then spraying the aerosol onto the surface of the substrates, as shown in Figure 5. The aerosol jet printing technique permits working with a large range of viscosities (0.7–2500 mPa·s) and lateral resolution even below 10 μm [117].
was achieved with 30 wt.% of silver content. Kell et al. [123] developed a screen-printable ink what seemed suitable for screen printing and generated lines with the resistivity of 5.5×10⁻⁵ Ω·m. The electrical resistivity of the printed silver traces is 2.7 times that of bulk silver.

3.1. Formulation of Silver Inks for Inkjet Printing

Shen et al. [118] prepared highly stable dispersions of Ag NP inks and printed them on paper and polyethylene terephthalate (PET) substrates using an inkjet printer. The low resistivity (3.7 μΩ·cm) of the printed silver patterns can be achieved when the sintering temperature is 180 °C. Shahariar et al. [119] successfully produced particle-free silver ink by selecting a silver salt mixture with an amine solution, which can be converted into silver particles once heated to a temperature when a silver ion can be reduced. The conductive tracks are created by inkjet printing on uncoated polyester textile knit, woven, and nonwoven fabrics without changing the feel, texture, durability, and mechanical behavior of the material. However, the packing and tightness of fabric structures and fiber sizes of fabrics highly impact the conductivity and resolution of the printed tracks. Zope et al. [120] developed a solid silver complex for formulating silver ink by using silver oxalate as a precursor and ethylenediamine as a complexing agent. This solid silver complex provides longer shelf-life stability. In addition, a hybrid thermal-photonic curing approach was presented, which increased the electrical properties and substrate adhesion. The electrical resistivity of the printed silver traces is 2.7 times that of bulk silver.

Hyun et al. [121], for instance, used Ag NPs-based inks to print conductivity lines by screen printing where poly(acrylic acid) (PAA) acted as a capping agent and ethylene glycol as solvent. The synthesized silver ink was prepared with a high loaded concentration (77 wt.%) what seemed suitable for screen printing and generated lines with the resistivity of 5.5×10⁻⁴ Ω·cm. Wang et al. [122] synthesized Ag NP inks in an aqueous solution and printed the inks on cotton fabric using screen printing. Low resistivity (2×10⁻⁵ Ω·m) was achieved with 30 wt.% of silver content. Kell et al. [123] developed a screen-printable ink containing metal carboxylate salt as a silver metal precursor and a polymer binder as the carrier. That screen-printed traces with good electrical (12 μΩ·cm) and mechanical properties were achieved using thermal or photonic sintering. The addition of ethylcellulose to silver precursor formulation increases the viscosity of ink to enable screen printing, lowers the decomposition temperature of the silver neodecanoate salt, and improves trace adhesion to the plastic substrate, leading to robust mechanical properties.

Apart from the requirements regarding the Ag NPs or the silver precursors, the different types of inks should be adequate for the processing type. From those, inkjet, screen, and aerosol jet printings are the more relevant and are discussed below, as shown in Figure 5.

**Figure 5.** Schematic of inkjet printing, screen printing and aerosol jet printing.

Typically, the inks for inkjet printing require a viscosity in the range from 0.001 to 0.1 Pa·s and surface tension from 15 to 25 mN·m⁻¹ [28]. Therefore, during and after the Ag NPs synthesis must be taken care of fitting these recommended ranges. That is usually assured by controlling Ag NPs concentration, using a suitable solvent or additives. Cheon...
et al. [124] reported on the synthesis of Ag NPs for inkjet printing using electrolysis. The authors showed that they could obtain Ag NPs of different sizes (10–80 nm) by controlling the applied voltage and the electrolyte temperature. The used electrolyte consisted of citric acid, hydrazine monohydrate, dispersion agent in deionized water. Kosmala et al. [125] reported on the synthesis of aqueous Ag NPs inks by a simple wet chemistry method, resulting in the formation of Ag NPs with a diameter around 50 nm. The authors used a triblock copolymer as Ag NPs dispersing agent and a high-intensity focus ultrasound to reduce the particle size from ~200 nm to 50 nm. Chen et al. [126] used a one-step synthesis of Ag NPs in a toluene solution containing dodecanoic acid or octadecanoic acid with the addition of n-butylamine and hydrazine, as shown in Figure 6. The Ag-dodecanoic acid NPs were finally suspended at 10 wt.% in cyclohexane to be employed in inkjet printing. Ag NPs in deionized water. In addition, 25 vol.% of isopropanol was used to adjust the surface tension and viscosity of the ink. Conductive patterns were obtained by inkjet printing the Ag NPs inks onto photo paper. Low resistivity was achieved with sintering at 200 °C.

![Figure 6](image-url)

**Figure 6.** (a) Schematic illustration of the whole process of Ag NPs synthesis; and (b) the direct printing process.

Jung et al. [127] showed a simple way of synthesizing and sintering Ag NPs employing a simple thermal decomposition process. The Ag NPs were synthesized by simple heating and stirring of AgNO₃ in the presence of the surfactant (oleylamine), without a reducing agent. After dispersion and centrifugation in toluene and methanol, Ag NPs were dispersed in hexadecane at the concentration of 30 wt.%. Tung et al. [128] synthesized Ag NPs with different shapes (spherical, prism, rod, and multifaceted NPs) by variation of PVP concentration using a rapid water radiolysis approach via X-ray diffraction analysis (XRD). The resulting NPs aspect ratio from 1 up to ~50 was reached for optimized reagent concentrations. The ink concentration for inkjet printing was set to 15 wt.% in methanol. Novara et al. [129] used “in situ” reduction of silver by printing AgNO₃ solutions on the porous silicon surface covered with reactive hydrides. The authors also investigated the influence of AgNO₃ concentration and solvents (water-ethanol, water-dimethyl sulphoxide) on the morphology of the printed Ag NPs layer. Zhang et al. [130] used a tannic acid (TA) to
stabilize the synthesized Ag NPs, which resulted in an environmentally friendly approach to upscale the inkjet printable Ag NPs based inks. The Ag NPs were prepared by the employment of the silver ammonia as a silver source and TA as a reducing and capping agent simultaneously. The whole reaction was carried out in a deionized water medium at room temperature for a very short time (30 min). The formation of Ag NPs was observed by the color changes of the solution color from colorless to yellowish and finally dark brown. Subsequently, the Ag NPs with an average diameter of 15 nm were purified in ethanol and deionized water, and collected by precipitation and drying. The Ag NPs could be stored without oxidation for several months. Ag NPs inks at the solid content of 1–10 wt.% were prepared by dispersing different quantities of Ag NPs in deionized water. In addition, 25 vol.% of isopropanol was used to adjust the surface tension and viscosity of the ink. Conductive patterns were obtained by inkjet printing the Ag NPs inks onto photo paper. Low resistivity was achieved with sintering at 200 °C.

Liu et al. [131] showed how to synthesize bimodal Ag NPs by changing the solution temperature and concentration of AgNO₃. The authors first synthesized Ag NPs of 10 nm in size at room temperature and AgNO₃ concentration of 100 g·L⁻¹, and subsequently, larger Ag NPs of 50 nm at a boiled state and AgNO₃ concentration of 0.54 g·L⁻¹. Finally, the prepared Ag NPs of the two different sizes were mixed in different ratios. The ratio of 2:1 (Ag NPs of 10 nm in size and Ag NPs of 50 nm in size) resulted in a decreased resistivity of 3.66 × 10⁻⁶ Ω·cm, with respect to each of them individually. Furthermore, the improved mechanical stability of the printed layers was also observed for this ratio.

Barrera et al. [132] synthesized Ag NPs by employing two different methods (chemical and microwave radiation). The chemical method was based on adding AgNO₃ into a solution of NaBH₄ and subsequently adding sodium citrate and PVP to stabilize the solution. The synthesis by microwave radiation was realized by mixing DMF, AgNO₃, and PVP in the tube of a single-mode reactor microwave under constant magnetic agitation. The Ag NPs prepared by microwave radiation were mixed with polyvinyl butyral (PVB), resulting in a nanocomposite with improved antibacterial properties with respect to Ag NPs prepared by chemical method.

Trinh et al. [133] prepared Ag NPs by chemical reduction of AgNO₃ using NaBH₄ with chitosan (CS) and cetyltrimethylammonium bromide (CTAB) as capping agents. The synthesized Ag NPs improved ink stability (more than 6 months) and cubic crystal structure with a size under 10 nm. After the synthesis, Ag NPs were mixed with glycerol, isopropanol, and deionized water to achieve the ink with appropriate viscosity and surface tension.

Another synthesis approach was used by Hao et al. [134], who applied a carboxyl-terminated hyperbranched polymer (CHBP) to stabilize the synthesized Ag NPs according to the procedure shown in Figure 7. The Ag NPs with almost monodisperse particles and a diameter of 10-20 nm were synthesized by chemical reduction of AgNO₃ using NaBH₄ in an aqueous phase. The final resistivity of the printed Ag NPs layers sintered at 180 °C was 10.83 μΩ·cm, which is approximately seven times higher than in case of bulk silver (1.58 μΩ·cm).

Yang et al. [135] showed a simple way to prepare Ag particle-free inks, which could be a possible cheap substitute for Ag NPs based inks, as shown in Figure 8. The ink was prepared by simple dissolution of a silver oxalate powder in different alcohols (methanol, ethanol, butanol, hexanol and 2-methoxyethanol or a mixture of them) using 1,2-diaminopropane (1,2-DAP) as the ligand. After the printing and sintering at 180 °C, the resulting silver layer resistivity was 15.46 μΩ·cm, which is only ten times higher than that of bulk silver.
3.2. Formulation of Silver Inks for Screen Printing

The screen printing technique typically works with viscosities ranging from 0.5 to 5 Pa·s and the surface tension from 38 to 47 mN·m⁻¹ [28]. Therefore, to comply with such requirements, Ag NPs need to be dispersed in a suitable polymer matrix and solvent, which assure the sufficiently high viscosity of the ink. Jadav et al. [136] used the bottom-up technique to synthesize Ag NPs, which were subsequently used to formulate a silver/silver chloride (Ag/AgCl) screen-printable ink for fabrication of electrodes for detection of vitamin C concentration from fruit juices. The authors used epoxy resin as a binder combined with a commercial hardener to achieve the desirable rheologic characteristics that made it suitable for the screen printing process. In another work of Liu et al. [137], the authors used an anion-mediated synthesis to prepare monodisperse Ag NPs. They used AgNO₃ as a metal source, in combination with glucose, PVP, and different anions (–SO₄²⁻, –PO₄³⁻, –CO₃²⁻ and –Br⁻) to tailor the morphology of the particles. The authors also investigated the influence of the nitrate-glucose ratio and reaction temperature on the shape and size of the synthesized Ag NPs. These nanoparticles were then re-dispersed in ethanol at a concentration of 30 wt.%, subsequently screen-printed onto the PET and paper substrate, and sintered at room temperature. Ding et al. [138] used a one-step polyol method to upscale the Ag NPs synthesis in ethylene glycol in the presence of PVP. The particle size (from 52 to 120 nm) was controlled by the mass ratio variation of AgNO₃ and PVP. Subsequently, the Ag NPs were re-dispersed in ethanol at 70 wt.% and printed.
in different conductive structures on PET substrate, reaching conductivities close to bulk silver, as shown in Figure 9.

**Figure 9.** Schematic illustration of the whole process of screen printing Ag NP-based ink preparation from the synthesis to the printing of conductive pattern. (a) The preparation of Ag NPs with different size distribution; (b) The dispersion of Ag NPs in ethanol to form silver inks; (c) Screen printing of silver inks. Reprinted from Ref. [138].

### 3.3. Formulation of Silver Inks for Aerosol Jet Printing

The aerosol jet printing technique permits working with a large range of viscosities (0.7–2500 mPa s) and lateral resolution even below 10 µm [117]. Salary et al. [117] also point out that aerosol jet printing is very sensitive to ink temperature and solvent evaporation rate, which may cause instabilities during the printing, such as pre-drying or particle accumulation within the printer nozzle.

Shankar et al. [139] used the non-aqueous synthesis in toluene with dodecylamine, and subsequent addition of silver acetate and a small amount of tin acetate, resulting in a formation of Ag NPs of 5–20 nm in size. The synthesized Ag NPs were then dispersed in decane and the viscosity (1–2 mPa s) of the solution was adjusted by adding 2-butoxyethanol at the concentration of 20 % by weight. Ivanov et al. [140] used a pulsed-periodic gas discharge in an air atmosphere to produce Ag NPs with a multigap gas-discharge generator from 99.95% purity silver. The resulting Ag NPs were then deposited by a focused aerosol beam on a glass substrate. Sonawane et al. [141] deposited branched polyethylenimine (BPEI)-functionalized Ag NPs on a substrate utilizing aerosol with flowing argon gas. BPEI-functionalized Ag NPs were dispersed at a concentration of 0.02 mg/mL to create the aerosol, the solution was poured into an ultrasonic nebulizer with an additional inlet for argon gas and an outlet connected to a quartz tube, followed by the plasma treatment. After the plasma treatment, the size of the Ag NPs was reduced from 10 to 5 nm, and a well-dispersed film was formed. The plasma treatment resulted in morphological changes in Ag NPs, enhancing their optical properties. Additionally, the plasma treatment resulted in a blue shift of approximately 70 nm in the peak of the surface plasmon resonance, which is typical for the high-temperature thermal treatment. Therefore, no additional thermal sintering was necessary in this case. The desired deposition and plasma treatment can be applied to a wide range of nanoparticle systems on flexible substrates.

### 4. Post-Printing Treatments

In order to form effective conductive paths, sintering processes are commonly necessary after the deposition of conductive silver inks, especially in metal-based nanoparticles inks. Generally, Ag NPs are dispersed in an aqueous/organic solution capped with organic stabilizers that act as an insulator, affecting the conductivity of printed tracks. Sintering removes the organic components by heating to the desired temperature, as shown in Figure 10. The sintering treatment induces the removal of stabilizers and coalescence of solid nanoparticles, and the sintering temperature is mainly dependent on the type of stabilizers to remove [142]. For reactive Ag inks, in addition to the organic solvent evaporation, the sintering process triggers chemical reactions in the ink to produce conductive silver patterns. Understanding the correlation between sintering temperature, sintering
process type and time and the morphology of nanomaterials, and the electrical properties of printed patterns is important to predict and improve the performance of the ink. There are many sintering methods, such as conventional thermal sintering, chemical sintering, electrical sintering, photonic sintering, and plasma sintering.

4.1. Thermal Sintering Method

Thermal sintering is the most straightforward method as the temperature removes the stabilizers either by evaporation or decomposition. For reactive silver inks, the sintering temperature triggers the chemical reaction of the silver precursor. Therefore, the thermal sintering highly depends on the size of silver particles, the volatilization or decomposition temperature of organic compositions in the inks, and the boiling of the solvent [143].

The effects of sintering temperature and time on microstructure and electrical properties of silver ink films have been investigated by Yang et al. [144]. Sintering at higher temperatures leads to a larger particle size of silver nanocrystals in printed film. The morphology of the films changes with the sintering temperature, resulting from the fast evaporation of the solvent and thermal decomposition and reduction of the complex. Moreover, with increasing time, the printed films show relatively more compact microstructures consisting of small silver particles with better film uniformity.

The high disadvantage of this method is that the temperature required for thermal sintering is not easy to implement on many substrates, such as plastic, paper, or elastomers. Hence, the development of low-temperature sintering technologies is particularly relevant in this field.

4.2. Chemical Sintering Method

Chemical sintering can be carried out at room temperature by adding chemical reagents, which can cause destabilization of the metal particle/stabilizer system and allow direct contact of the metal nanoparticles to form conductive paths. Among chemical sintering, two possible methodologies, i.e., dissolving or detachment of the protective layers on surfaces of nanoparticles, can be applied [28,142,145,146].

As a dissolving method, Wakuda et al. [147,148] studied the extraction of the dodecyl-lamine stabilizing molecules from nanoparticles by dipping printed lines in several alcohols (ethanol, methanol, isopropanol). Dodecylamine has excellent solubility in alcohols, which can remove the weak bonds between Ag NPs and dodecylamine, causing coalescence and sintering of Ag NPs. An example of a detachment method is the one presented by Grouchko et al. [149], where chloride caused room temperature sintering of Ag NPs stabilized by polyacrylic acid sodium salt. As illustrated in Figure 11, due to the presence of
the destabilizing agent, the desorption of the stabilizing polymer from the surface of the nanoparticles occurs, causing coalescence and sintering of Ag NPs. In that study, about 41% of the conductivity of bulk silver could be reached by using hydrochloric acid (HCl) vapors as a sintering agent. This method makes the possibility to print on heat-sensitive materials.

![Figure 11. Schematic illustration of chemical sintering of nanoparticles due to the detachment of stabilizers.](image1)

### 4.3. Electrical Sintering Method

Electrical sintering consists of applying a current across the printed pattern resulting in local heating due to the highly resistive nature of silver ink before sintering [142,143,150], as shown in Figure 12. Consequently, electrical sintering is a local Joule heating sintering method, which minimizes the thermal damage to the substrate [151]. Electrical sintering can tailor the degree of sintering by controlling the heating current. Hummelgård et al. [152] investigated the entire sintering process in real-time using an in situ TEM. The onset of sintering and coalescence of nanoparticles starts at power levels of 0.1–10 mW/µm³. In addition, when the ink was Joule-heated, the first cause of conductivity increase was the carbonization of the solvent. Instead of burning off the solvent, carbonization helps prevent micro-cracking of the sintered structure.

![Figure 12. Schematic illustration of an electrical sintering setup.](image2)

The temperature affects the specific resistance of printed patterns during the electrical sintering process. Moon et al. [153] studied the relationship between the specific resistance of the ink and the sintering temperature using the Wiedemann–Franz law showing that the resistance of the ink decreases as the process temperature rises. However, thermal damage can result from the high initial resistance of printed patterns. Stepwise current electrical sintering was devised by Lee et al. [154]. By increasing the current gradually, the conductive line can endure a higher current since the specific resistance has progressively dropped during the process. Compared with the constant current-supplying electrical sintering
method, the enhanced final-step current produces lower resistance of the conductive line without damaging printed patterns.

4.4. Photonic Sintering Method

The sintering of printed metal patterns by electromagnetic irradiation ranging between the ultra-violet (UV) and infrared (IR) is called photonic sintering [155]. Metal nanoparticles on common substrates such as polymer foils or glass show very intense absorption in the visible region due to their plasmonic properties. There is a significant difference in the optical properties between the metal nanoparticles to be sintered and common substrates. Glass and other common polymer substrates have absorption only in the UV range, except PI because of its brown color. Under an appropriated emission spectrum and sufficient optical power density, photonic sintering exploits the possibility of heating the printed ink by photothermal effects, leaving the transparent substrate material unaffected [8,142]. Photonic sintering includes laser sintering, IR sintering, and intense pulsed light (IPL) or photonic flash sintering.

The laser sintering is performed using a high-power laser as the power source to melt and fuse the powders into solid structures. The energy provided by laser is absorbed by printed tracks in a selected area controlled by a computer system, resulting in decomposition of organic components in the inks and rapid sintering. The nanoparticle inks heat up due to the photothermal effect and then are sintered due to the locally elevated temperature. After laser sintering, the parts with desired electricity and functionality can be obtained [143]. The generated temperature inside the ink has to be controlled and kept as low as possible to avoid the dissipated heat from the ink to the substrate, which can be realized by tuning the pulse length, flashing frequency, sintering time, and intensity of the lamp. Hong et al. [156] developed a method to fabricate a metallic grid transparent conductor on a plastic substrate using selective laser sintering of metal nanoparticles. The fine grid conductor yields a high transmittance (>85%) and low sheet resistance (<30 Ω/sq), which can be directly applied to a large-area flexible substrate. Theodorakos et al. [157] compared three different laser sources with different pulse duration (continuous wave, pulsed nanosecond and picosecond) at 532 and 1064 nm to sinter Ag NP inks. It was found that continuous wave and nanosecond-pulsed laser sintered the NPs inks efficiently but may damage the substrate depending on the laser power and the spatial laser intensity profile. Alshammari et al. [158] found a strong dependence of the morphology and electrical performance of the printed samples on the process parameters, namely the laser fluence and the scanning speed of the laser. The received conductivity values with optimal laser condition at a scan speed of 5 mm/s and beam energy of 30 mJ/cm² are comparable to that obtained for the thermally sintered ones, which is about one order of magnitudes less than that of bulk silver. The printed conductive patterns were then successfully utilized as electrodes for organic thin-film transistors.

IR technology uses irradiation in the range of NIR to MIR region (700 to 15,000 nm) to dry and sinter printed inks within a short time. Moreover, Sowade et al. [159] reported roll-to-roll (R2R) IR drying and sintering of printed Ag patterns on flexible polymer substrates with high web velocities (up to 1 m·s⁻¹) within one second. The obtained results contributed to the development of IR system to sinter materials for R2R-printed electronics. The IR conditions, including the power of the IR lamp, the distance between the IR lamp and substrate, and the sintering time, should be tuned appropriately to avoid the damage of the flexible substrate and printed patterns. Park et al. [160] performed IR sintering in a large-area R2R gravure printing process for printed Ag NPs and Ag flake layers. Relevant process conditions, e.g., IR lamp power, distance from the IR lamp to printed patterns, and exposure time, were varied to evaluate the effects on the sheet resistance of the two types of pastes. They found that the distance between the substrate and IR lamp is the most influential parameter in all the samples, whereas the second most effective parameter is the sintering time. The lamp power is insignificant in the Ag NP pattern, but it is significant in the Ag flake layer.
IPL employs a Xenon flash lamp with a wide wavelength ranging from hundreds to thousands of nm to generate heat in the target materials to achieve the necessary temperature increase, as shown in Figure 13. However, high-intensity light onto the front surface of metal nanoparticle patterns often causes the defects, such as delamination, cavities, and cracks in the patterns. Kwak et al. [161] presented a back-irradiation method for obtaining defect-free high conductivity Ag NP patterns in a few milliseconds, wherein the back surface of the patterns is irradiated with IPL through a transparent plastic substrate. In contrast to the back-irradiated patterns, the vapor is trapped in the front-irradiated patterns. It delaminates the patterns from the substrate because the front of the patterns acts as a barrier to vapor venting.

![Figure 13. Scheme of the IPL sintering setup. A Xenon flash lamp with a wide wavelength is used to generate heat in the target materials.](image)

4.5. Plasma Sintering Method

In general, plasma sintering is performed by exposure of printed patterns to low-pressure argon plasma. During plasma generation, the plasma produces excited species, such as ions, radicals, and UV irradiation, causing decomposition of the stabilizing agent and enabling direct contact between metal particles [162]. The plasma sintering process starts from the top to the bottom in the structure, and sintering does not occur with greater thickness than the penetration depth of the plasma species [163]. High conductivity values of up to 60% of bulk silver were obtained by Perelaer et al. at a temperature compatible with the glass transition temperature of common polymer foils using two sequential sintering techniques, low-pressure argon plasma sintering and microwave flash sintering [164]. Instead of low-pressure plasma sintering, Wünscher et al. [165] combined atmospheric pressure plasma sintering and a mild thermal treatment to reduce the sintering time and improve the conductivity of inkjet-printed Ag NP inks. Temperature-assisted atmospheric pressure plasma sintering revealed a conductivity of 16% of the bulk silver and reduced the process time by a factor of five compared to low-pressure plasma sintering as well as conventional thermal sintering. Ma et al. [166] sintered Ag NP inks on glass substrates using argon plasma and studied the effects of plasma parameters on the electrical property, material composition, and film nanostructure of sintered Ag NPs films. The lowest resistivity of the sintered layer was five times higher than that of bulk silver.

5. Outlooks and Perspectives

Ag NPs have attracted considerable attention due to their high electric conductivity, good oxidation resistance, optical characteristics, and antibacterial effects. As follows from the above overview, many kinds of nanoparticles can be synthesized, which could be utilized in electronic, optoelectronic, and biological fields. Ag-based inks can be printed on various substrates to obtain conductive patterns. The strategies of achieving highly conductive Ag NPs-based designs by moderate sintering have been given in this review. A better understanding of the relationship among the sintering condition, the protective agents, Ag NPs size, and shapes has been covered. The development of fast, efficient, and inexpensive sintering methods is also highlighted.
Although remarkable scientific progress in the preparation of nanoparticles and sintering of conductive patterns has been achieved, there are still challenges that call for further development in order to be implemented by the industry at a large scale. The current high price of commercial Ag NPs is one of the critical factors. The development of new Ag NPs deserves more attention, which could decrease the ink dosage and printed ink thickness and increase the electric conductivity. In addition, selecting a suitable sintering method for the printing patterns is crucial to obtain good conductive printed patterns. An unsuitable sintering method could cause higher resistance and porosity after the metal ink is sintered. Since the temperature-sensitive substrates (flexible plastic and paper) require low-temperature sintering, further development of new sintering techniques, which are compatible with R2R printing processes, is crucial to their industry applications.

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