Insight into the fabrication and perspective of dendritic Ag nanostructures

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ABSTRACT
This article reviews recent advances in the shape-controlled synthesis routes of hierarchical Ag dendritic nanostructures. Various fabricated techniques, with or without templates or surfactants, electrochemical method and sonochemical, microwave or photochemical assisted synthesis method, can be amazingly effective in the dendritic Ag nanostructures synthesis, if synthetic parameters are carefully selected. The involved synthesis mechanism are discussed in order to control the limited factors along the fabricated progress efficiently and then to find new synthetic means. Also, the blemish of the above synthesis routes that hinders the practical application is pointed out to inspire the future research.

KEYWORDS
Ag dendrites; synthetic technique; DLA model; oriented attachment

1. Introduction
Recently, there has been a tremendous attention in the design and fabrication of noble metal nanostructures of complex morphologies due to their prominent anti-bacterial [1–3], catalytic [4,5] and optical properties [6,7]. Among these noble metals nanostructures, silver nanostructures with super-hydrophobic surface and narrow plasmon resonance inspired majorities of researchers to investigate. Researchers have found that the fascinating properties of metal nanostructures strongly depend on the involved particle size, the inter-particle distance and the shape of nanoparticles. It is very important to control the size, shape and distribution of the Ag nanoparticles (AgNPs) to systematically investigate their antibacterial, catalytic and electro-optical properties and new applications. Numerous of synthetic routes has been developed for fabricating various Ag NPs, such as wires [8,9], rods [10,11], triangular plates [12], cubes [13–15], polyhedrons [16], dendrites [17,18], etc. Dendritic Ag NPs have hierarchical branches and leaves as shown in Figure 1. It stands out among other nanostructures due to its large surface area and its unique interfacial structure towards adding substrate or metals, which bring its amazing application in surface-enhanced Raman scattering (SERS) [19,20], surface-enhanced fluorescence [21], super-hydrophobic [22–24], antibacterial [25,26], chemical sensors [27,28] and catalysis [29–31].
Ag dendrites are usually formed under non-equilibrium conditions by controlled driving force, while strict driving force control is usually difficult [32]. To obtain uniform morphology and high crystalline quality, many physical and chemical techniques have been developed. Generally, Ag dendrites synthetic technique can be divided into four kinds: self-structure confinement method, template method, physical chemistry method and soft solution method. The self-structure confinement method can directly fabricate Ag dendritic nanostructures only under very limited conditions. The template method and soft solution method can well regulate Ag fractals growth but it is not good to the purity of dendrites. The physical chemistry methods including electrochemistry, photo-chemistry, sonochemistry, microwave, etc. are considered as effective routes, but they usually require special equipment and higher cost. This review briefly summarises the advantages and challenges of different synthetic techniques. More importantly, shape-controlled mechanism of Ag dendritic nanostructure is discussed as well.

2. Fabrication of dendritic Ag nanostructures

2.1. Ag dendrites fabrication via direct growth

Simple replacement reaction or reduction reaction without template or surfactant for Ag nanostructures formation has been widely applied. Active metals and other reducing agents are mostly used for Ag dendrites formation. The reaction process just as a simple galvanic displacement, metals just transfer electrons to reduce Ag⁺ ions, and then Ag NPs deposited on the surface of metals, as displayed in Figure 2. Among these, metals like Cu [33], Mg [34], Zn [35], Fe [36] and Sn [37] have been reported in many researches. Copper is widely used due to its higher standard reduction potential, which makes reaction process easy to control and no copper oxides exists in the final products. The single-crystalline dendrite with the overall length of about 200 nm was fabricated by reducing Ag⁺ in the AgNO₃ solution on Cu mesh [33], while the length of silver nanodendrites

Figure 1. Typical SEM image of Ag fractal dendrites.
formed on Cu rods is about 3–8 μm [38]. The factors such as the concentration of silver ions and reaction time have significant influence on Ag nanostructures. Low concentration of AgNO₃ could lead to incomplete morphologies of Ag nanostructures, and higher concentration of Ag⁺ would promote the growth of nano-silver particles [39]. With increasing the reaction time, the size of Ag nanoparticles increased and the structures of Ag dendrites became clearer. Finally, a high-quality single-crystal dendrite was formed.

In addition, different anions such as SO₄²⁻, NO₃⁻ and CH₃COO⁻ in silver nitrate also influence the structures of silver dendrites which are potentially useful in SERS. The SO₄²⁻ ions can speed up the growth of Ag dendrites which the crystal growth direction is preferentially oriented to one direction by comparison of nitrate ions [40]. Cai et al. used a wet chemical fabrication approach to form leafy spike-like silver dendrites by directly immersing Cu into 0.06 M CH₃COOAg solution for 10 h at 48 °C, and the aggregated flakes of Ag nanostructures is about 20–80 nm in thickness [41]. The results also revealed that the concentration of silver ions, reaction time and temperature could also affect the Ag dendrites formation.

Compared with copper, metals like iron, zinc, tin and magnesium are more active and more easily to reduce silver ions to form dendrites. Silver dendrites formed on the surface of Zn foil grow along a preferential direction in the replacement reaction process [35]. A similar growth trend of Ag dendrites which have a diameter distribution from ~20 to 90 nm also occurred on tin plate reacting with AgNO₃ solution [37]. Mg reacts with AgNO₃ solution can form single-crystal Ag dendrites, which grow along a preferential direction at the beginning is similar to the other replacement reaction and shows a good uniformity. Besides, Mg is often used to reduce silver ions together with other metal ions simultaneously to get a variety of nanostructure alloy. The bimetallic dendrites such as Ag/Cu, Ag/Pd, Au/Pd, Cu/Au, Cu/Pt and Cu/Pd have great prospects in many areas. The main stems of Ag/Cu bimetallic dendrites formed by immersing Mg into silver nitrate and copper sulphate mixed solution were typically 20–30 μm, and the first-level branches of the dendrites were 1–10 μm [34]. Nersisyan et al. used Fe instead of Mg to fabricate Ag/Cu bimetallic dendrites at room temperature [36].

In addition to the active metals, other reducing agents also have been widely used for Ag dendrites fabrication. Silver ions were reduced to silver atoms and the silver atoms gathered into a ball at the beginning of the reaction process. Finally, the Ag dendrites

Figure 2. The schematic galvanic displacement of tin reacting with Ag⁺ [37].
formed with the reaction time increasing [42]. Ascorbic acid (AA) as a weak reductant provides a way to form significantly branched Ag particles from the reduction of AgNO$_3$ sometimes [43–45].

### 2.2. Ag dendrites fabrication via template method

Although the method without template is simple, the process control is more difficult. In order to control the reaction process properly, many Ag dendrites were fabricated via template methods. The templates usually include surfactants, metals or non-metals with specific structures and polymer films. Among these, the surfactants have been widely used, such as tetraoctylammonium bromide (TOAB) [46], cetyltrimethylammonium bromide (CTAB) [47] and polyvinyl pyrrolidone (PVP) [48]. PVP is a common water-soluble macromolecule and PVP chains can not only adsorb the silver nanoparticles, which is beneficial for silver nanoparticles growing along a specific direction, but also can act as a weak reductant to reduce silver ions. The ratio of PVP to AgNO$_3$ has great influence on the structure of Ag dendrites, by adjusting the ratio of PVP/AgNO$_3$, Ag NPs with different morphology can be formed [48]. During the reaction, silver ions accepted electrons and Ag NPs formed rapidly. The silver isotropic particles contact with one another more easily to form dendrites on the condition of higher concentration of AgNO$_3$ [49]. Chiang et al. [50] adjusted the concentration of PVP to form the dendritic-shaped and needle-shaped Ag/Cu alloy particles, which simultaneously coated on copper surface. The results showed that the ratio of Ag/Cu affected the structure of particles. The higher concentration of PVP would lead to smaller dendritic-shape and lower thermal conductivity. Zhu et al. [51] proposed that the surfactant PVP and the concentration of soluble species would change the surface chemistry of Ag crystal. Both factors have influence on the dendritic crystal growth behaviour. The TOAB, CTAB and other surfactants acted similar as PVP, but different surfactants would have different reaction conditions. Agrawal et al. [46] reported that the Au/Ag alloy nanostructures can be formed at low concentration of TOAB and CTAB. In addition, the surfactants were key factors for the formation of Ag dendrites [47].

Besides surfactants, metals or non-metals with specific structures such as silicon [52], nickel [53] and anodic aluminium oxide (AAO) [54] were mostly used in the Ag dendrites fabrication. The self-assembled silver nanostructures grew in template by ion track technology, and the template coated with active sites could support the formation of Ag nanoparticles. The self-assembled Ag dendrites formed in porous Si/SiO$_2$ matrix and SiO$_2$ nanoparticles didn’t need any additional reagents during the reaction [55]. Factors that affect the Ag nanostructures formation in the templates include the structure of the template, presence of surface active agents, and the reaction conditions. Under different conditions, silver nanostructures assembled in porous Si/SiO$_2$ matrix have different shapes such as ‘sunflowers’, ‘azalea’ or ‘corn’. With the presence of CTAB, the SiO$_2$ particles interact with the CTAB head groups through electrostatic interaction initially, and then Ag$^+$ react with Br$^-$ from CTAB to form the AgBr/SiO$_2$ nanoparticles. Finally, the small Ag nanoparticles can move freely into the aqueous solution and gathered onto the Ag dendrites [56]. Sol-gel method can also be used to synthesise the silver oligomers in a SiO$_2$ amorphous matrix [57].
Furthermore, the self-assembled Ag dendrites could be directly formed on the polymer template such as poly-n-butylacrylate-co-(acrylonitrile-co-2-methacrylic acid 3-(bis-carboxymethylamino)-2-hydroxy propyl ester) (PBAGI) [58]. Figure 3 reflects the process of Ag dendrites growth process on PBAGI [27]. In this process, Ni NPs served as reducing agents assisted with PVP acted as capping agents. Ag would be formed on Ni NPs via galvanic replacement when Ag⁺ collided with Ni NPs in the solution firstly, and then like polyol process, Ag NPs bounded by PVP in the bulk solution would diffuse towards the as-grown NPs on the PBAGI membrane and form the self-assembled Ag dendrites. In addition, Gong et al. [59] reported the fabrication of silver halide NPs in template Ag(I)-coordinated Langmuir–Schaefer (LS) films. The LS films without long alkyl chains were helpful for the diffusion of nanoparticles and made it possible to form larger aggregations of nanostructured materials. Dendritic Ag nanocrystals with highly ordered structures can also been formed in aqueous PEO–PPO–PEO tri-block copolymer (F127) solution [60]. The morphology is directly determined by not only the concentration ratio between reductants AA and F127 but also the F127 induced kinetics.

Along with the in-depth study, some novel templates are applied to the synthesis of silver nanoparticles such as the cellulose dialysis cassettes [61] and macroporous jellyfish mesoglea [62]. The physical–chemical properties of novel templates affected the structure and properties of silver nanostructures. However, the manufacturing process is cumbersome and expensive compared to the other templates. Table 1 summarises the Ag dendrites synthesised by template methods, including detailed reaction conditions and various templates.

### 2.3. Ag dendrites fabrication via electrochemical method

Electrochemical method is a versatile, rapid and cost-effective technique for the preparation of nanostructured materials. The morphology of silver NPs depends on the distance
from the synthesis conditions to thermodynamic equilibrium. And the driving force and interaction of interfacial dynamics of electro-crystallisation exactly provides a condition of far-from-equilibrium, which is beneficial for hierarchical dendritic Ag nanostructures [63,64]. The electrodeposition process can be assisted by surfactants or templates, which have impacts on the morphology and size. Dendritic silver nanostructures can be manufactured in the presence of a suitable surfactant. Fu et al. [65] fabricated Ag dendrites/AgCl hybrid thin film on ITO electrode with electrolyte consisting of silver-ammonia \( \text{Ag(NH}_3\text{)}_2\text{OH} \) and certain amount of hexadecyl trimethyl ammonium chloride \( \text{CTAC} \). Investigation revealed that the Ag dendrites consisted of a long central stem with side branches emerge at about 55° and when using higher content of CTAC, the growth of Ag dendrites was suppressed while the numbers of polyhedral AgCl dramatically increased. Polyethylene glycol (PEG) [66], mixed surfactants of cetyltrimethylammonium bromide (CTAB) and sodium dodecyl sulphate [67] and sodium dodecanesulphonate (SDS) [68] are also mixed with AgNO₃ solution for fabrication of dendritic silver nanocrystals. The individual Ag dendrites on conductive glass had 2D structures with one trunk (long axis) and branches (short axes). They were obtained at static potentials between 1.5 and 2.7 V with assistance of PEG [66]. PEG also plays an important role in controlling the growth morphology of the silver particles on glassy carbon electrode (GC), dendritic Ag NPs can only be formed with higher concentration of PEG [69]. While in AgNO₃–SDS aqueous solution, silver dendrites were obtained on titanium substrate [68].

In terms of pulsed ultrasound that is often used to prepare silver NPs [70], surfactant or organic capping reagents are often needed in the shape-selective synthesis of NPs, because they control the growth of crystals by selectively adhering to certain crystallographic planes. When introducing ultrasound to electrochemistry, mono-disperse dendritic nanostructure can be synthesised. The sonoelectrochemical technique is using a current pulse to form massive nucleation, followed by a burst of ultrasonic pulse to remove the deposit from the electrode before the next current pulse. PVP is commonly utilised in sonoelectrochemical technique, which can be thoroughly washed out after synthesis [71]. PVP also affects the morphology of Ag NPs with different electrolyte solution.

| Templates | Ag precursor | Reaction conditions | Reference |
|-----------|--------------|---------------------|-----------|
| Surfactant | AgNO₃ | 70 °C, 2 h | [49] |
| Polyvinyl pyrrolidone (PVP) | AgNO₃ | r.t., 2 h | [46] |
| Cetyltrimethylammonium bromide (CTAB) | AgNO₃ | 80 °C, 3 h | [47] |
| Tetraoctylammonium bromide (TOAB) | AgNO₃ | Ultrasonic, r.t., 15 min | [54] |
| Metals and non-metals with specific structures | AgNO₃ | r.t., 30 min | [56] |
| Silicon | AgNO₃ | 120 °C, 6 h | [52] |
| Raney nickel | AgNO₃ | r.t., 2 h | [53] |
| Anodic aluminium oxide (AAO) | AgNO₃ | Ultrasound, r.t., 15 min | [54] |
| Si/SiO₂ | AgNO₃ | r.t., 30 min | [56] |
| Polymer | AgNO₃ | In dark, 25 °C, 7 days | [58] |
| PBAGI | AgNO₃ | r.t., 3 min | [59] |
| Langmuir–Schaefer (LS) films | AgNO₃ | r.t., 24 h | [60] |
| Aqueous PEO–PPO–PEO tri-block copolymer (F127) | AgNO₃ | r.t., 36 h | [62] |
| Novel templates | AgNO₃ | Stirring, 1 h | [61] |
| Cellulose dialysis cassettes | AgNO₃ | r.t., 1 h | [61] |
| Macroporous jellyfish mesoglea | AgNO₃ | r.t., 36 h | [62] |

a) r.t. = room temperature.
When using a saturated solution of silver citrate, the prepared silver NPs can only agglomerated to be dendritic crystals without PVP owing to the dissociation equilibrium of \( \text{Ag}_3\text{C}_6\text{H}_5\text{O}_7 \) [70]. While in \( \text{AgClO}_4 \) solution, it is worth to mention that PVP favours the formation of side branches and leaves and promotes the formation of finer and more hierarchical nanostructures [72]. This is because PVP selectively adheres to different crystallographic planes on the surfaces of NPs to enable anisotropic crystal growth. DNA [73] and nitrilotriacetate (NTA) [74] also affect the fabrication of Ag dendrites by sonochemical technique. Investigation revealed that the dispersion property of DNA enhanced the stability of silver dendrites because of the interaction between DNA and the silver particles [73]. And ultrasound helped removing the Ag particles from the cathode, cleaning the cathode surface and replenishing the double layer with Ag ions. The shape of Ag NPs is extremely influenced by the concentration of NTA. Increasing the concentration of NTA to a certain content in the synthetic system is found to be favourable for the formation of the shaped particles, especially for Ag dendrites [74].

Besides, uniform dendritic silver NPs can also be synthesised by coupling electrodeposition and template method. A proper template agent can easily control the size and shape of the desired Ag hierarchical crystallisation, while the applied potential or current can be used to accelerate Ag nucleation and Ag ions transportation. Polymers are commonly utilised as templates in dendritic Ag NPs synthesised by electrodeposition. A study showed the synthesis of highly crystalline silver NPs in \( \text{AgNO}_3 \) solution with the assistance of triblock copolymer P123 as the soft template. It has been found that the concentration of P123 has a great impact on the morphology of Ag NPs. When the electrolyte contains 0.1 M \( \text{AgNO}_3 \) with 2 wt% P123, multilevel dendritic structures are clearly observed; and when the concentration of P123 reached to 5 wt%, the deposit showed a cabbage-like morphology. In contrast, Ag NPs without any well-defined shape were obtained without P123 [75]. Besides the polymer template in electrochemical fabrication of dendritic Ag NPs, carbon material can also be a good template. Li et al. [76] investigated the aid of graphene oxide (GO) or PSS modified multi-wall carbon nanotubes (MWCNTs) for silver dendritic nanostructures synthesis on ITO, well-defined Ag dendritic structures with a central stem length of approximately 5 \( \mu \text{m} \) were obtained. This is owing to the linear structure and negative surface charges which could attract positively charged Ag\textsuperscript{+} as the nucleation sites for Ag NPs.

Although template or surfactant are beneficial to the synthesis of dendritic Ag NPs, external additives not only make the synthetic procedure complicated and introduce impurities but also may release toxic chemicals to the environment. The Ag dendrites fabrication without any template, surfactant or stabiliser by electrodeposition is getting more and more popular. Jing et al. [77] reported electrochemical preparation of Ag dendrites with long central backbone on aluminium slice, which could be used as active and stable SERS substrate. Investigations revealed that Ag source concentration, electrolyte, applied potential or current density and deposition time all played significant roles in Ag dendrites formation. Different synthesis conditions are shown in Table 2. A higher concentration of Ag source resulted in a denser dendritic Ag. When the Ag\textsuperscript{+} concentration is high enough that ions diffusion is not a limiting factor, Ag dendrites just agglomerate to polyhedral shapes [78–80]. The morphology of silver structures was found to be highly dependent on the electrolyte concentration. Sharma et al. [81] noted that when the concentration of \( \text{KNO}_3 \) increased (lower than 15 mM), the dendritic Ag NPs became
more pronounced with the size changing from 0.5 to ca. 20 μm. Lower applied potential with longer deposition time led to successful formation of hierarchical Ag dendrites [79,82]. Also, a suitable applied current density is in favour of Ag dendrites synthesis under the same deposition time. In a case study, when the applied current is lower than 200 μA, only Ag rods with majority of protrusions are found, which look like the precursor of branches (as shown in Figure 4(A,B)). While increasing the current density from 300 to 500 μA, Ag dendrites are largely synthesised with coarse branches and leaves (Figure 4(C,D)). Further increasing the current density from 700 to 2000 μA, Ag dendrites become thinner and longer, and finally grow as cubic particles (Figure 4(E–G)).

Table 2. Summary of recent Ag dendrites prepared by electrodeposition without surfactant or template.

| Ag precursor | Working electrode | Current or voltage | Deposition time | Reference |
|--------------|-------------------|--------------------|-----------------|-----------|
| 0.01–0.05 M AgNO₃ | ITO | 5–15 mA | 12 s | [78] |
| 1–30 mM AgNO₃ | Aluminium rod | 25–2000 μA | 5–300 s | [80] |
| 1–20 mM AgNO₃ and 0.1 M KNO₃ | Au foil | (0–0.6) V | (10–240) s | [79] |
| 10 mM AgNO₃ and 0.1 M KNO₃ | ITO | -0.10–0.50 V | 3 min | [83] |
| 5 mM AgNO₃ and 0.1 M KNO₃ | ITO | (-0.2–0.8) V | (1–10) min | [82] |
| 0.01 M AgNO₃ | Ni/Cu film | (-0.4–2.0) V | 30 min | [16] |
| 1 g/L AgNO₃ and 0.51 g/L KNO₃ | Microwell patterned ITO | -0.9 V | (2.5–10) min | [63] |

Figure 4. SEM images of the Ag NPs synthesised at various applied current densities for 10 min of reaction. Current density increase from 25–2000 μA (A–G) [80].
Besides DC current or potential for Ag dendrites synthesis, square-wave pulse electrochemical method has also been investigated. The square-wave potential and its frequency determine the formation of the dendritic structure [84]. In a case study, Ag dendrites were deposited on carbon fiber by applying the pulse potential. The morphology changed from dendrite to plate structure with the square-wave pulse from low frequency to high frequency [85]. Pavlović et al. [86] analysed the effects of current or overpotential wave shapes on the morphology of Ag NPs. It was found that in square-wave pulsating overpotential regime, the particles are less dendritic and less agglomerated than the ones obtained in the constant regime. Because of the demand for higher power and a high-speed potential stat by using pulsating overpotential deposition and the lower-quality of Ag dendrites forming under this condition, DC potential or current deposition are developed more popular.

2.4. Ag dendrites fabrication via other methods

2.4.1. Sonochemical synthesis

Sonochemical processing has proved to be an effective technique for generating novel materials with remarkable chemical and physical properties. Researches have shown that acoustic cavitation is the key factor influencing the reaction efficiency. By the action of ultrasound, the formation, growth and implosive collapse of bubbles occur in the liquid medium. The collapse of bubbles generates localised hot spots with transient temperature of about 5000 K, pressure of about 1000 atm, and cooling rate of over 10^9 K/s [87]. Such specific condition, to a large extent, facilitates the far-from equilibrium growth of metal nanostructures dramatically [72]. Ultrasound has been known as an effective method in shape-controlled synthesis of silver nanostructures for some time now.

The most prominent feature of introducing ultrasound to synthesis is the acceleration of mass transport and reaction rates. Xiao et al. [53] reported the synthesis of silver dendritic nanostructures using Raney nickel template with the assistance of ultrasonic waves. Wang et al. [87] developed a preparation of well-defined silver dendritic nanostructures from AgNO₃ aqueous solution in the presence of isopropanol and PEG by using ultrasonic irradiation without any template. Generally, at the beginning of the reaction, only silver spherical nanoparticles were obtained. Then the silver nanoparticles aggregated together to assemble silver nanodendrites. Defined silver nanodendrites were observed with proper control the sonication time. The dendritic nanostructures would transform to compact crystals if they are irradiated for an overlong period.

Gao et al. [88] prepared silver hierarchical dendrites via a simple integration method using Devarda’s template as a reducing agent and architecture template in aqueous system. In this system, the ultrasound irradiation plays an important role in dendritic nanostructures fabrication. It is proved that ultrasonic waves can accelerate the diffusion of Ag⁺ into the channels of the Devarda’s templates, which leads to the formation of well-defined nanoscaled silver dendrites.

2.4.2. Microwave-assisted synthesis

As a widely used heating method, microwave irradiation shows its advantages in conducting and accelerating chemical reactions because of the rapid and selective heating characteristics. Microwave-assisted method became a preferred technique in chemical synthesis
in the last dozen years or so, especially in materials synthesis. Generally speaking, there are two main mechanisms involved in this process: the dipolar mechanism and the electrical conductor mechanism [89].

Some recent studies reported microwaves as the energy supply in the formation of Ag dendrite. He et al. [90] reported the formation of silver dendritic nanostructures in DMF (N,N-dimethyl formamide) with PVP as the stabiliser and the assistance of microwave irradiation. It has been proved that microwave irradiation could induce the faceted growth of silver crystal and the formation of the anisotropic dendritic silver capped by PVP. Noroozi et al. reported that dendritic silver nanoparticles could also be successfully fabricated by similar way without reducing agents [91]. Compared to conventional heating method, the silver nanoparticles suspension produced by microwaves was much more stable.

2.4.3. Photochemical method

Light is another frequently used driving force in metal nanostructure synthesis. Zhou et al. reported a novel ultraviolet irradiation photoreduction technique to prepare single-crystal Ag nanorods and dendritic nanostructures at room temperature using polyvinyl alcohol (PVA) as a protecting agent [92]. Zou et al. developed a new photochemical seed-mediated method to synthesise silver nanostructures with 253.7 nm UV light in the presence of PVP [93]. It demonstrated that silver dendrites were obtained only at the place exposed to UV irradiation. For a given condition, irradiating the solution with UV light for an excess time was not helpful as dendrites would change into thermodynamically more stable nanocrystals.

Reagent-free nanostructure synthesis is one of the key hot spot aspects nowadays. Ahmed et al. used electrostatic hybrid films of porphyrin—polyoxometalate (POM) for photocatalytic synthesis of silver dendrites [94]. Porphyrins can be excited by visible light which play the role of photosensitisers. Electrons were transferred to POM, which is known as a good catalyst to reduce silver ions. In this system, giant silver dendrites have been obtained. Nie et al. synthesised silver dendritic structures on a wide band gap GaN epitaxial thin film, which could produce vast numbers of reductive electrons under continuous UV irradiation [95]. In the study, uniform spheroidal nanocubes were firstly formed on the planar GaN surface, and then grew into Ag dendrites with hyper-branches.

3. Growth mechanisms of dendritic Ag nanoparticles

The phenomenon of dendritic crystal growth is a particularly elegant example of a process in which a simple system under a non-equilibrium condition generates complex but highly structured new patterns. The well-known growth mechanism under non-equilibrium conditions has been explained on the basis of a diffusion limited aggregation (DLA) model [96] and oriented attachment growth [97,98]. Controlling the driving force under far-equilibrium conditions is extremely difficult, while a kinetic regime could be created by the balance of diffusion and reaction rates for the growth of dendrites. The DLA model can well explain the fractal phenomena in the non-equilibrium reactions [96]. The formation mechanism of Ag dendrites is related to the angles fractal dimension ($D$), the radius between a number of arbitrary aggregate sites being covered by a Hausdorff set ($r$), that is, the non-vacant boxer size and the total number of non-vacant boxes ($N(r)$), which can be defined as

$$N(r) \sim r^{-D}$$
where $D$ can be calculated with a box-counting algorithm [99]. After plotting $\log(N(r))$ vs. $\log(1/r)$, the value of $D$ can be calculated from the slope. As shown in Figure 5, $\log(N(r))$ vs. $\log(1/r)$ is well fitted by a straight line with a slope of ca. 1.83, which is close to the fractal dimension (ca. 1.7) of a DLA model. Besides, it can also indicate that the dendritic nanostructures exhibit good symmetry and self-similarity characteristic of fractals.

It is undeniable that the diffusion of Ag precursors plays an important role in the formation of the Ag dendrite nanostructures. Cluster formation occurs by adhering particles together with a random path to a certain seed in contact and then these particles form a growing structure. Surfactant or template control the nuclear and directional aggregate which give rise to a non-equilibrium system thereby in favour of forming Ag dendrites [101,102]. When dendritic Ag NPs were prepared at the solid/liquid interfaces via electroless deposition under Langmuir monolayers [103], it is clear a DLA process. The solid/liquid interface resulted in a higher rate of galvanic reduction, and this rapid reduction of Ag$^+$ ions led to fast nucleation and growth of Ag NPs and different diffusion rate of the Ag$^+$ ions in the interface and sub phase. After Ag$^+$ ions reached the pre-formed NPs, the bulge part of the Ag NPs contacted the ions and grew for Ag dendrites formation. Microwave irradiation and ultrasonic waves can also control the diffusion kinetics of Ag NPs to grow from cluster to Ag dendrites [90,104]. When Ag dendrites are formed with a suitable template under microwave or ultrasonic waves, the adhesive Ag particles tend to form clusters with random path to a selected seed on contact. A typical example to explain this mechanism is electrochemical synthesis of Ag dendrites [64]. The diffusional mass transport of Ag precursor can be regulated by the viscosity of the solvent, such as ethanol, ethylene glycol and glycerinum [105,106]. Figure 6 shows the relationship between diffusion coefficient of Ag$^+$ and the morphology of Ag dendrites. With the increase of diffusivity, the Ag dendritic structures become longer, and the angles between branches and trunks are larger than that with lower diffusivity. Fast diffusion gives chances for all facets to receive silver atoms in Ag dendrites growth; while under slower diffusion conditions, the silver atoms for dendrite structures growth are limited and these silver atoms prefer to move at the high energy facets to minimise the surface free energy. The formation mechanism of Ag dendritic NPs synthesised by electroless metal deposition is schematically illustrated in Figure 7, where the silicon etching and Ag deposition occur simultaneously [107]. This synchronous growth process is considered within the DLA model. At initial stage, the concentration of Ag precursor and reductant is high, which leads to the reduction-nucleation-growth of Ag nanoclusters, and particles diffuse

Figure 5. Double logarithmic plots of $N(r)$ vs. $r$ for the Ag dendrites [100].
and stick to the formed nanoclusters subsequently. As the deposition proceeding, both Ag precursor and reductant concentration decreases and the nanoclusters surface energy decreases, too. The low surface energy drives the Ag nanoclusters to form dendritic Ag nanostructures.

Different from DLA model that starts from primary building blocks like atoms, ions or molecules, oriented attachment model spontaneously arranges adjacent primary Ag NPs rather than Ag ions with common crystallographic orientation while attaching, followed by joining these particles at a planar interface [108, 109]. The primary Ag NPs are free to move and rearrange for the low-energy structure. Usually, this mechanism can illustrate the formation of single-crystalline Ag dendrites under hydrothermal conditions [110] or chemical reduction [111, 112]. However, when the reductants

Figure 6. SEM images (A–H) of Ag particles synthesised at different volume ratios of glycerol in water under 500 $\mu$A cm$^{-2}$ [105].
are foil, rod or plate immersing into Ag precursor solution, Ag will nucleate and grow on the substrates with no free crystallography unless using suspended micro-particles as reductant. As reported by Fang et al. [113], Ag dendrites were synthesised on the zinc plate in replacement reaction. Two aggregates share the same crystallographic orientation and then the polycrystalline aggregates transform to single-crystalline Ag dendrites after the self-assembling process. The driving force for spontaneous oriented attachment is the elimination of high-energy surface, which will lead to virtual decrease of the surface energy, while Ag dendrite has a considerably increased surface energy in contrast to the equilibrium shape, such as the hexagonal structure. As the reaction proceeds, the concentration of Ag$^+$ ions drop to a certain level and then the small grains has enough time to grow to the thermodynamically favoured shapes, instead of dendrites. As Ag dendrites formation is a time-dependent process, sometimes, the growth of Ag dendrites may follow both DLA model and oriented attachment. Mdluli et al. [114] studied the time dependent evolution of Ag nanodendrites with PVP as surfactant. Formamide reduced Ag$^+$ to form anisotropic Ag particles, and PVP adsorbed Ag ions firstly and then formed a weak confinement interaction with anisotropic nanoparticles which influenced the gradual growth of Ag NPs to further oriented nanodendrites. The presence of surfactant not only allows diffusion of new particles into the surface of the existing particles followed with formation of new tree-like structures, but also minimises the surface energy for nanoparticles oriented attachment [109].

Figure 7. Schematic illustration of the growth process of the Si nanowires and Ag dendrites [107].
4. Outlooks

Well-defined Ag dendrites by different synthetic routes and the possible growth mechanisms have been summarised in this review. The amazing hierarchical structure of dendritic Ag nanostructures shows great potential in catalysis, antibacterial, super-hydrophobicity, optical enhancement as well as sensors for detecting glucose, hydrogen and diazepam etc. Among these applications, enhanced SERS activity has been widely investigated because the branches and tips of Ag dendrites have strong surface plasmon polarisation. Nowadays, electrocatalysis of Ag dendrites becomes another research hotspot because of its excellent sensitivity as sensors. There is no doubt that the function of Ag dendrites in many fields offers a potentially rewarding opportunity for forming other metal nano dendrites. Nevertheless, regardless of the assistance of surfactant, template or capping agents for Ag dendrites formation, or the assistance of electrical field, microwave, ultrasonic waves and light, every synthetic route has its own superiority or drawbacks. The self-structure confinement method has the advantages of low cost, high yield and easiness of control, but the synthetic kinetic is affected by temperature or reductants and it may cause pollution. The template method can regulate the growth process and produce well aligned dendritic Ag arrays with uniform size, but the template cost is high and its separation from Ag dendrites may complicate the synthetic process and reduce the product purity. Besides, the particle size and shape can be effectively controlled via the assistant of ultrasound, microwave or light. However, these extra assistance would increase the energy consumption and need additional production facilities. Similarly, electrochemical method is also a rapid and environmental friendly technology for dendritic Ag NPs synthesis and it can control the growth kinetics and particle size well with additional electricity input. Hence, environmental friendly and energy saving synthesis method should be investigated. Furthermore, much effort should be done for fabricating Ag dendrites with precise size control, high purity and excellent yields. In brief, the fabrication and application of dendritic Ag NPs also launched a new world for studying other metal nanoparticles.

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