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SOLAR IRRADIATION AND *NAGEIA NAGI* EXTRACT ASSISTED RAPID SYNTHESIS OF SILVER NANOPARTICLES AND THEIR ANTIBACTERIAL ACTIVITY

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Silver nanoparticles have gained great attention from engineers and scientists due to their vast applications in modern nanodevices. In the present investigation, silver nanoparticles were synthesized from a fast and environmentally friendly pathway by using leaf extracts of *Nageia Nagi*, which acted as a fast reducing and stabilizing agent. When the silver ions exposed to the *Nageia Nagi* leaves extract, the silver nanoparticles were bio-reduced within 25 seconds in the presence of sunlight. They stabilized in few minutes resulting in green AgNPs without an external source of heating, they showed efficient antimicrobial behavior against multi-drug resistant bacteria Escherichia coli and Staphylococcus aureus using Kirby–Bauer method. Synthesized silver nanoparticles were characterized by Ultraviolet–Visible (UV–vis) Spectrometer, Transmission Electron Microscopy (TEM) and Energy Dispersive X-ray Analysis (EDAX).

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1. Introduction

Nanotechnology is a promising and rapidly growing field for its vast applications in different science and technology sectors [1]. For years, noble metal nanoparticles like gold, platinum and silver are widely used in medicinal applications, among those silver nanoparticles have attracted great attention of researchers in the last two decades. The surface Plasmon resonance and large effective scattering cross section of individual silver nanoparticles make them ideal candidates for molecular labeling where phenomena such as surface enhanced Raman scattering (SERS) can be exploited [2]. Biological pathways for the synthesis of silver nanoparticles have gained great attention from the engineers and scientists explaining the mechanism of the synthesis. The novel plants based fabrication of nanomaterials has a wide range of applications including antimicrobial properties. Biosynthesis from plant extract based silver nanoparticles with controlled physicochemical properties has been reported, such as Citrus sinesis, Geranium and Aloe Vera etc (Table 1). Silver nanoparticles prepared using biological materials has the properties of a high surface area, smaller in size and high dispersion. These prepared nanomaterials have many applications, including spectrally selective coating for solar energy absorption, generation of intercalation materials [3].

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Table 1. Different novel plants used for the synthesis of silver nanoparticles with their particle size.

S. No.	Novel Plants	Particle Size	Reference
1	Acalypha indica	20–30 nm	[7]
2	Citrus sinensis	3–12 nm	[8]
3	Geranium	16–40 nm	[9]
4	Aloe vera	15 ± 4 nm	[10]
5	Mangifera indica	~20 nm	[11]
6	Medicago sativa	86–108 nm	[12]
7	Aloe ferox	5 nm	[13]
8	Allium cepa	33.6 nm	[14]
9	Andrachnea chordifolia	3.4 nm	[15]
10	Phyllostachys aurea	13± 3.5 nm	[16]
11	Ilex Cornuta	7–20 nm	[17]

However, reaction times of most biological synthesis methods of silver nanoparticles are slow, and reduction of 1mM silver nitrate requires mostly about 24-120 hours, which restricts the industrial practice of synthesis methods [4]. Visible light has been shown to increase the synthesis rate of AgNPs, and solar radiation is effective in gold nanoparticles synthesis [5, 6]. In addition, the use of natural sunlight would reduce energy costs.

Generally, sunlight in chemical processes is considered to be a non-polluting, non-toxic and traceless facet [18-20]. In spite of many sunlight advantages, its uses are limited due to seasonal and regional reasons. This investigation engages with a quick synthesis of silver nanoparticles assisted by *Nageia Nagi* leaf extract in the presence of sunlight and its antibacterial efficiency (Figure 1). The plant (Nageia Nagi) used in this study, is a hardy tree species, can withstand a range of weather conditions. Nageia Nagi is a dioecious tree belonging to Podocarpaceae plant family native to mainland China. Its wood has been used for building houses and furniture for years, the leaves and seed oil is usually used for cooking purposes in China [21].

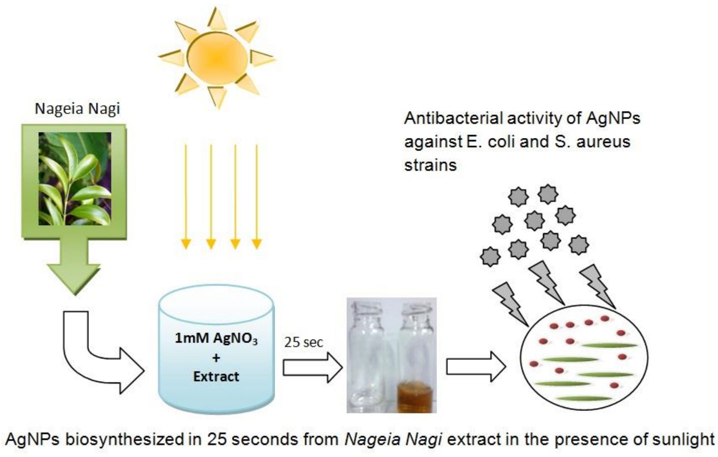

Fig 1. Schemetic illustration of AgNPs biosynthesis

3. Materials and Methods

3.1 Plants and Chemicals

The *Nageia Nagi* leaves were collected from their trees available in the Xiasha campus of Zhejiang Sci-Tech University, Hangzhou, China. Silver nitrate ($AgNO_3$) was purchased from Strem Chemicals, Inc. (USA). In this study, fresh leaf extract and distilled water were used throughout the experiments.

3.2 Preparation of the extract

Fresh leaves of Nageia Nagi were collected, thoroughly washed with double distilled water and cut into small pieces. Dried 5 g of finely cut Nageia Nagi leaves were weighed and boiled at $100^{o}C$ for 10 minutes in 250 ml beaker containing 100 ml of double distilled water. The extract obtained was stored at $4^{o}C$ for further use.

3.3 Synthesis of silver nanoparticles

In this experiment, one conical flask containing 5ml of freshly prepared Nageia Nagi leaves extract with 5ml of 1mM aqueous $AgNO_3$ solution was kept in a dark place, having 90-80 ^{o}C temperature as is taken off from the boiler, while another conical flask with same materials was kept under the sunlight (having 90-80^{o}C temperature as taken off from the boiler). The silver ions were reduced to silver nanoparticles within 2 minutes by the *Nageia Nagi* leaves extract kept in a dark place. A quick conversion of silver nanoparticles was achieved in 25 seconds only with *Nageia Nagi* leaves extract in the presence of sunlight. The formation of silver nanoparticles was confirmed by the rapid change of solution color from colorless to yellowish-brown in both cases.

3.4 Antibacterial assay

Silver nanoparticles biosynthesized from Nageia Nagi extract was tested for antimicrobial activity by Kirby–Bauer method against pathogenic bacteria Escherichia coli (Gram-negative) and Staphylococcus aureus (Gram-positive). The pure bacteria cultures were sub cultured on nutrient agar media. Both strains were swabbed evenly onto the single plates using sterile glass rods. After incubation at $37^{o}C$ for 24 hours, the levels of zone diameter inhibition of bacteria were measured.

4. Results and Discussions

Silver nanoparticles were synthesized with an eco-friendly process without any synthetic chemicals like reducing agents or stabilizers. Generally, AgNPs are followed to reduce silver ions to silver nanoparticles from different methods, such as; Ultra Violet or Gamma radiation, prolonged reflux, photochemical, ultrasound and by some toxic chemicals. These methods are either very expensive or chemically toxic effecting environment somehow. Therefore, the main purpose of this research has been to produce silver nanoparticles without any harmful chemicals relatively using just natural eco-friendly leaves.

During the reaction of $AgNO_3$ with freshly prepared Nageia Nagi leaves extract, it was observed that the color of the solution was changed to dark yellowish brown in a few minutes. It is well known that silver nanoparticles exhibit yellowish brown color when synthesized, as the color change was due to excitation of surface Plasmon vibrations in the metal silver nanoparticles. From Figure 2, it was observed in UV-visible spectrum that surface Plasmon resonance occurred in between 400-450 nm indicating the clear formation of silver nanoparticles in the solution in both of the cases (sunlight and dark).

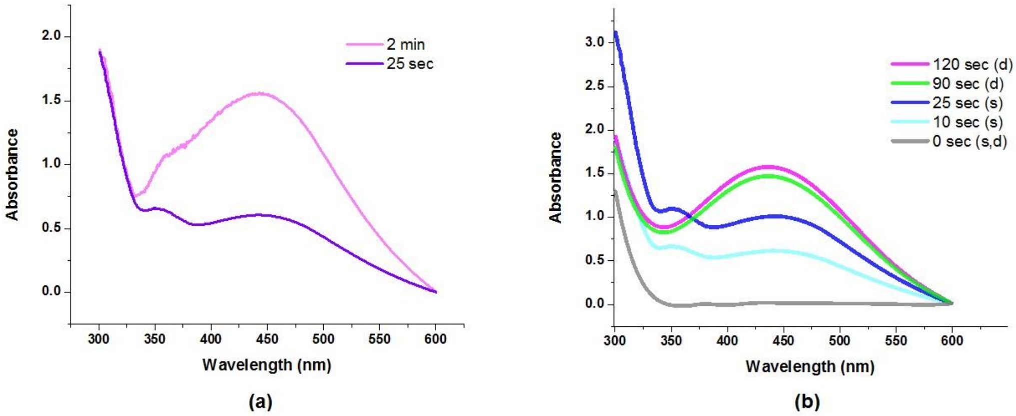

Fig 2. UV-visible spectrum of AgNPs biosynthesized by solar irradiation (a) rapid AgNPs synthesis in th presense of sun light and (b) various AgNPs curves synthesised in sunlight (s) and dark (d)

Study of various reaction times in the reduction process revealed that the synthesis of silver nanoparticles starts within seconds of time with Nageia Nagi extracts in the presence of bright sunlight and reaches the plateau after a few minutes if kept in the dark (Fig. 2a). In the first few seconds, an exponential increase in absorbance was observed, which remains constant yet increasing the time of exposure of samples in sunlight. The absorbance values flatten out after 2 minutes in the case of sample kept in the dark. Incubation at different exposure times to the sunlight resulted in the same major peak, which indicates that similar particle sizes and shapes of AgNPs were produced in both cases (Fig. 2b). Accordingly, suggests a very fast kinetics reaction of nanoparticles with the help of Nageia Nagi extract and sunlight irradiation.

The formation of silver salts to silver nanoparticles is possible by different methods, such as some of the natural and synthetic hydrophilic polymers having functional groups like hydroxyl (OH), carboxylic (COOH), amino (NH2), or thiol (SH) groups offers reduction and as well as stabilizing abilities to the formed nanoparticles. It also has been reported that sunlight can be used as a reducing agent for the synthesis of silver nanoparticles, several studies showed possibilities of reduction of silver nanoparticles from sunlight (photo-reduction) [22-23].

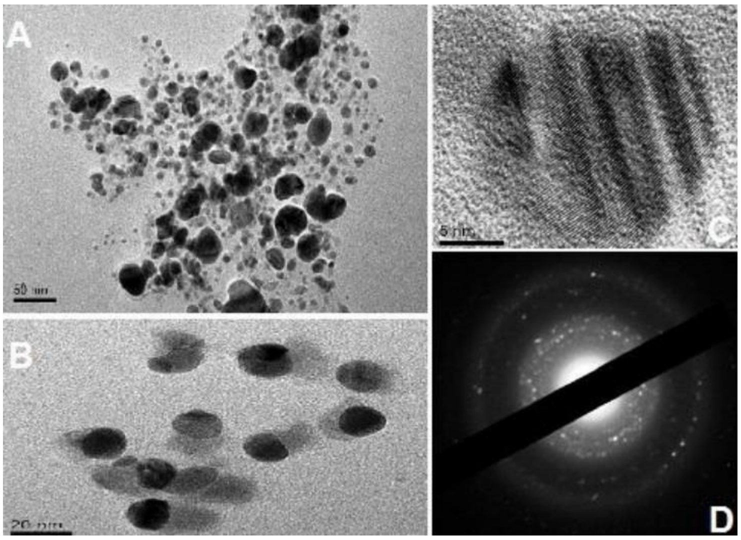

Fig 3. TEM micrograph of the silver nanoparticles: (A) The scale bar correspond to 50 nm, (B) The scale bar correspond to 20 nm, (C) The scale bar correspond to 5 nm and (D) shows the SAED pattern of corresponding AgNPs

The TEM examination of prepared silver nanoparticles revealed the presence of poly-dispersed spherical nanoparticles of varying sizes (Figure 3). These shapes are common in AgNPs synthesized by plant extracts. The diameter distribution of 100 AgNPs was in the range from 3.8 to 25.9 nm with a mean size of 16.1 nm (Fig. 4a). EDS spectrum confirmed the presence of intensive absorption of silver nano-crystallites (about 3 keV) (Fig. 4b).

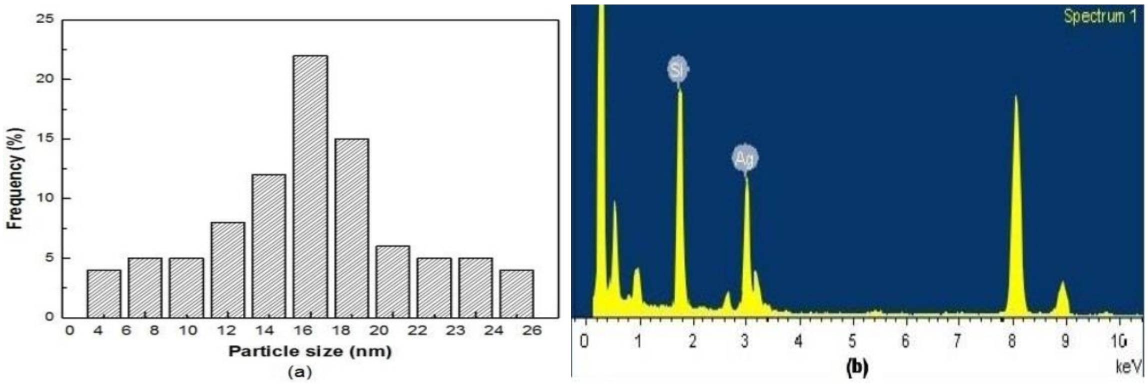

Fig. 4 (a) Histogram of the nanosilver produced and (b) EDX spectra of AgNPs

The antibacterial activity was achieved by measuring the diameter of the zone of inhibition. Figure 5 shows the inhibition by Nageia Nagi extract assisted AgNPs towards two challenge bacteria. It was evident that around the paper soaked in AgNPs synthesized in the dark has a significant inhibition zone against E. coli and S. aureus was, with the mean diameter of the inhibition zone of 15 mm and 6 mm, respectively, however, there was nearly same inhibition for the AgNPs synthesized in sunlight against the challenge bacteria.

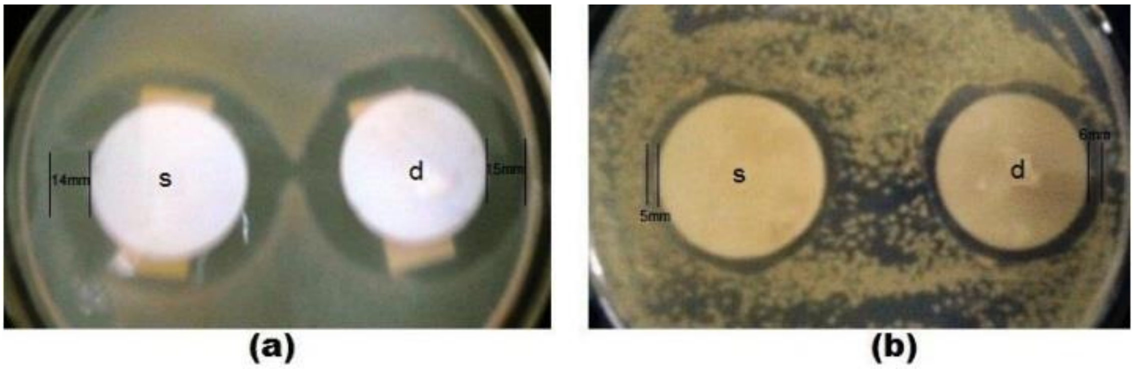

Fig 5. Bacterial growth inhibition against bacterial strains (a) E. coli and (b) S. aureus, by using silver nanoparticles synthesized in sunlight (s) and dark (d)

5. Conclusion

Today, nanotechnology symbolizes a new era of opportunities and applications to develop modern drugs based on synthesized nanoparticles having various biological properties. Shape and size are of synthesized silver nanoparticles are important for enhancing their antimicrobial activities. The synthesized silver nanoparticles using Nageia Nagi extract with and without the presence of sunlight demonstrated significant antimicrobial activities. The green synthesis of AgNPs emerged to be alternative to the chemical methods and can be suitable for developing an easy process industrial production. This study demonstrates a rapid and effective approach to synthesize silver nanoparticles from Nageia Nagi leaf extracts with a minimum reaction time of 25 seconds.

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