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Psidium guajava leaf extract-mediated synthesis of ZnO nanoparticles under different processing parameters for hydrophobic and antibacterial finishing over cotton fabrics

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ABSTRACT

In recent times, treatment of natural fibres, like cotton, with antimicrobial finishing using nanoparticles has become increasingly high due to the possibility of anticipated property and their tuneable nature. In this study, using the phytoconstituents of leaf extract of *Psidium guajava*, Zinc acetate was reduced to obtain ZnO nanoparticles using three different synthesis routes namely sonication, wet-chemical and hydrothermal methods. To evaluate and get a broader understanding about the influence of processing parameter over the ZnO nanoparticles, physico-chemical, biological and photocatalytic property, a comparative assessment was performed. The ZnO nanoparticles were subjected to X-ray diffraction, particle size analysis and scanning electron microscope to ascertain its structure/morphology. The comparative studies reveal that hydrothermal method of ZnO synthesis yields smaller particle size (12 nm), with higher photocatalytic activity (94%), higher antibacterial activity against *Escherichia coli* (27.4 mm) and *Staphylococcus aureus* (29.3 mm) and lower structural defects comparing to ZnO synthesised via other methods of synthesis. Furthermore, the most effective ZnO nanoparticles (hydothermal method) was incorporated in chitosan and coated over the cotton fabric to functionalise cotton fabrics for better antimicrobial and water repellent property. Along with the same line, the ZnO nanocomposite coated fabrics exhibit better hydrophobic (157 ± 0.1°) (superhydrophobicity) and higher antibacterial activity (30.58 ± 0.3 mm (99%) (*S. aureus*) and 24 ± 0.5 mm (96%) (*E. coli*)), which could be attributed to ZnO particle size and favourable structural morphology. Thus, the study not only reveals the tuneable nature of the ZnO nanoparticles with respect to processing parameters and also shows the potential role in coating these nanoparticles over the cotton fabrics to impart an effective antimicrobial and hydrophobic finishing for biomedical applications.

1. Introduction

Materials controlled to nano regime (i.e., particle size less than 100 nm) shows atom like behaviour due to the high surface to volume ratio and quantum size effect [1–3], leading to a critical and unique role in array of fields such as biomedical engineering [4,5], tissue regeneration [6,7], targeted drug delivery [3,8,9], textile [10,11], etc. Growing awareness of health and hygiene over the last decade has resulted in an exponential increase in the demand for treatment of natural fibres, like cottons, with antimicrobial finishing as it provides an immaculate growth medium for microbial growth due to its high ability to absorb sweat [12]. This ideal moisture conditions provided by the cotton fabrics, is responsible for the growth of microbes such as *Staphylococcus aureus* (*S. aureus*) and *Staphylococcus epidermis* (*S. epidermis*) which in turn, leads to skin infections such as boils, impetigo, cellulitis, and furuncles [11,13,14]. Along with the aforementioned fact, the growth of the microorganisms over the textile materials usher leads to discoloration and reduction in textile efficiency [10]. The meteoritic development in health, safety and environment legislations has resulted in a forever increasing demand for research and development in functionalisation of antibacterial property over technical textile materials such as sutures, bandages, gauze, surgical gowns and hospital linen [5]. To combat the aforementioned issues, researchers are trying to develop new antimicrobial finishing process with help of aldehydes, halogens,

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quaternary ammonium compounds and amines [15,16].

ZnO nanoparticles, due to its high photocatalitic activity [17], less cost, UV-shielding [12] and high antibacterial property [4] is attempted to use in textile industries. As reported elsewhere, ZnO nanoparticles shows high effective antibacterial activity against both gram positive and gram negative bacteria without the presence of light [1]. These nanoparticles, through different mechanisms like cell penetration, attachment on bacterial surface via electrostatic forces, and production of reactive oxygen species leads to inhibition of bacterial growth [6]. Therefore, ZnO nanoparticles show high potential for technological applications and can be effectively used for protection against antibacterial infection.

Traditionally ZnO nanoparticles are synthesised through solution method, i.e., liquid chemical reaction or liquid chemical reduction of zinc precursors such as zinc acetate, zinc nitrate, and other compounds using compounds containing OH functional groups, such as sodium hydroxide [18]. To meet the rapid demand of the technical textile market, industries use nanomaterials prepared through inorganic chemical routes to functionalise cotton fabrics with antibacterial property. Many synthesis processes are developed to meet this rapid demand in recent years to synthesise nanomaterials, such as Vapour-phase deposition [19], sol-gel [20], precipitation [21] and decomposition [22], etc., but this synthesis process produces nanoparticles with high degree of production cost along with it hazardous chemicals which are difficult to process are also released [22]. In order to overcome these issues, researchers focus on green synthesis i.e., green chemistry, a branch of Chemical engineering for the synthesis of materials with minimum use and generation of hazardous chemicals [12]. The branch specifically uses bio molecules to reduce the zinc precursors and provide pure ZnO nanoparticles with non-toxic by-products which can be easily processed [1].

Psidium guajava (*P. guajava*) or the common guava is an evergreen shrub or small tree belonging to the family *myrtaceae* [23]. *P. guajava* is used as a popular medicine against diarrhoea and is also used for wound dressing, ulcers, rheumatic pain [24,25]. *P. guajava* leaves are also being reported to show antibacterial, anti-inflammatory and anticancer properties [23,25]. *P. guajava* is selected for this study due to its high content of polyphenols (gallic acid, protocatechuic acid, caffeic acid, ferulic acid, chlorogenic acid, ellagic acid, guavin b), flavanoids (quercetin, leucocyanidin, kaempferol, quercetin3-β-l-arabinofuranoside), carotenoids (β-carotene, lutein, lycopene, β-cryptoxanthin, rubixanthin, criptoflavin, neochrome, phytofluene), triterpenes (oleanolic acd, ursolic acid, β-sitosterol, uvaol) present in the crude water extract of leaves [23]. The aforementioned biomolecules have functional groups that can coordinate Zn (II) and will help in stabilisation during the formation of ZnO nanoparticle [26]. The Major polyphenols i.e., Chlorogenic acid and Guavin-B present in the *P. guajava* leaf extract had being reported to show antioxidant activity [27]. Furthermore, Kaemferol and Leucocyanidin present as the major flavanoids and oleanolic acid and ursolic acid as the major titerpines in the *P. guajava* plant extract doesnot show or highlight any advantages with respect to chemical or biological attribute as per prior reports on the phytochemical studies on *P.* guajava plant extract [27]. β-carotene and Neochrome present in the *P. guajava* acts as a chain-breaking antioxidant and thus protects cell against photo-oxidant [27]. Apart from the aforementioned advantages of biomolecules present in *P. guajava* leaf extract, the leaf extract had being used to treat human inflammatory response, diarrhoea and also used as an analgesic and vasodiallitive medicines [27]. Therefore, *P. guajava* can be taken as an ideal biocatalyst/Chelating agent for the preparation of ZnO nanoparticles from zinc acetate precursor.

Recently, owing to its easy accessibility and low toxicity, researchers are trying to functionalise cotton fabrics with bionanocomposite i.e., components of natural polymer matrix mixed with organic/inorganic metal particles at nano scale [5,9]. As reported earlier studies, metal oxide nanoparticlres-chitosan nanocomposite is used in functionalising the cotton fabrics with antibacterial property and also in providing hydrophobic surface to reduce absorption of sweat [12]. Chitosan is being used in various industrial applications, due to its high antibacterial activity, high permeability, and non-toxic nature [5]. The effect of processing parameters on the formation of green synthesised ZnO nanostructures with optimised physico-chemical as well as biomedical properties is found to be scanty.

In this paper, different synthesis routes i.e., sonication, wet-chemical and hydrothermal methods are employed for the synthesis of ZnO nanoparticles using *P. guajava* plant extract. The resultant nanoparticles are compared and characterised for optical, morphological, and biological properties. The main objective of this study is to provide an excellent textural property by coating the optimised ZnO nanoparticles with respect to the processing parameters over the cotton fabrics. In addition, understanding the tuneable nature of green synthesised ZnO nanoparticles by change in the processing parameters and enhancement of functional properties of the cotton fabrics is one of the most fascinating emerging research fields for the upliftment of biomedical textiles.

2. Materials and method

2.1. Materials

The fresh leaves of *P. guajava* were collected from the fellow lands of Kolkata, West Bengal, India. The collected leaves were washed several times with double distilled water (DD) to eradicate any dust particles from the surface of the leaves, following which the leaves were shade-dried at room temperature for more than two weeks. The dried *P. guajava* leaves were subjected to ball milling for 15 h in order to get a fine powder. In order to prepare the leaf extract, 10 g fine powder of *P. guajava* nanoparticles were taken and mixed in 100 ml of DD water and kept undisturbed for a period of 24 h. Following which, the solution was taken and filtered using Whatman No. 1 filter paper to obtain the leaf extract of *P. guajava* for the green synthesis of ZnO nanoparticles.

2.2. Synthesis of ZnO nanoparticles

The ZnO nanoparticles were green synthesised using the leaf extract of *P. guajava* through three major synthesis routes namely i.e., Sonication [28], wet-chemical [29] and hydrothermal [30] methods. Synthesis parameters for the aforementioned synthesis routes were employed to a base solution. The base solution was prepared by adding 1 M zinc acetate precursor (Zn $(CH_3COO)_2$, an industrial grade from Merck, India) to 100 ml *P. guajava* leaf extract to synthesise ZnO nanoparticles. All the acquired AR grade chemicals were utilized without any further purification (Fig. 1). The sturcture of the major bioactive molecular comounds within the *P. guajava* natural extracts are shown in Fig. 1.

2.2.1. Sonication

Sonication at 40 KHz (Advanced sonicator, Lark, NEW Delhi, India) was employed over the prepared base solution for 1 h, till a clear suspension was obtained. Following the suspension of the precipitates in the base solution, the base solution was dried in a hot air oven at 80 °C for 12 h. The dried precipitate was then calcined at 600 °C for 2 h, to produce ZnO nanoparticles which were exploited for further physico-chemical characterisation studies. The obtained ZnO nanoparticles were hereafter termed as ZnO-s.

2.2.2. Wet-Chemical

For synthesising ZnO nanoparticles through wet-chemical route, the base solution was agitated vigorously using the magnetic pellet under elevated temperature (70 °C) for 1 h, till precipitate were formed. Then, the base solution was dried at 120 °C for 12 h, following which; it was calcined at 600 °C for 2 h. The calcined powder i.e., ZnO nanoparticle