A Review on Green Synthesis of ZnO Nanoparticles Using Coriandrum Sativum Leaf Extract For Degrading Dyes in Textile Wastewater: A Prospect Towards Green Chemistry

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Abstract. Nanotechnology involves material with nanoscale dimension that range from 0.1 to 100 nm, to make devices, systems and materials with essentially new characteristics and applications due to their large surface area to volume ratio. Zinc oxide nanoparticles (ZnO NPs) are widely known due to its wide band gap and high exciton binding energy. ZnO NPs are normally synthesized by chemical methods which involve the use of hazardous and expensive chemicals, resulting in toxic and environmentally hazardous by-products. Green synthesis of ZnO NPs by plants extract is non-toxic and economic. Therefore, this paper aims to explore the potential of plants extract in the green synthesis of ZnO NPs. Moreover, the capability of Coriandrum sativum leaf extracts to produce ZnO NPs in degrading dyes in textile wastewater is also discussed.

1. Introduction
The textile industry has been one of the most influential and highly demanded industries since the beginning of mankind. The manufacturing process of textile however includes the use of large amounts of water, dyes and chemicals to condition the textile to its specified requirements. These dyes and chemicals are composed of trace metals such as chromium, arsenic, copper and zinc that can be detrimental to both humans and the environment if left untreated [1]. In addition, it was found that approximately 10-15% of dyes utilized for the production of textiles are discharged with textile wastewater [2]. Commonly adopted water treatment methods that involve processes such as chemical flocculation and coagulation, sedimentation, aeration and filtration are ineffective in the case of removing dyes and chemicals from textile industry effluents [3]. Semiconductor photocatalysis is a method that has been developed for decades as an effective treatment for coloured effluents. This method involves the use of semiconductor photocatalysts and solar irradiation to catalyse the degradation and oxidation of contaminants into harmless by-products such CO2 and H2O. In comparison to conventional water treatment methods, photocatalysis has the advantage of being energy-efficient as it uses renewable solar energy. Other than that, photocatalysis is also found to be more feasible in terms of time-efficiency and the conditions required for its reaction [4]. Zinc oxide (ZnO) and Titanium dioxide (TiO2) are the most widely known semiconductor photocatalysts due to their relative affordability as well as their wide band gap energy resulting in their high oxidation ability [5]. However it was found that ZnO exhibited a higher efficiency in the degradation of dyes as...
compared to TiO2 and also serves as a more economical alternative in the long run [6,7]. Furthermore, ZnO has displayed higher photocatalytic ability in the form of nanoparticles (NPs) compared to bulk form. This is due to the increase in surface area of the ZnO NPs for the reaction to take place allowing for a higher rate of degradation of the dye [5]. ZnO NPs are commonly synthesised using unsustainable methods that involve the use of toxic chemicals and hazardous inert gases [8]. The toxic by-products resulting from the chemical synthesis of NPs also get adsorbed by the NPs which would then be released into the water body that is being treated and pollute the water source. As the by-products resulting from conventional chemical synthesis procedures are toxic, a more sustainable and environmentally-friendly method must be adopted. Biosynthesis, also known as green synthesis, is a method of synthesising NPs using plant extract [9]. This method produces bio-friendly NPs, does not involve the use of any hazardous chemicals and is adopted in the manufacturing of medicinal products [10]. Green synthesis also serves as a more economical alternative in comparison to chemical synthesis which is very expensive [11]. However, there only a few literature studies have emphasized the benefits of biosynthesis/green synthesis their implementations in the real industries. Thus, the main aim of this review was to review the green synthesis of ZnO NPs using Coriandrum sativum extract and its application in degrading the dyes in textile wastewater.

2. Potential of ZnO as a semiconductor photocatalyst
Photocatalytic degradation has been studied extensively due to the feasibility of the process being conducted in ambient conditions and using atmospheric oxygen as a source of oxidant [12]. ZnO and TiO2 have also been recorded in many occasions as inexpensive, benign, easily acquired and having the ability to completely mineralize organic contaminants into CO2, H2O and mineral acids [13]. Although TiO2 has exhibited higher efficiency in degrading cellulose bleaching effluents as well as organic dyes, higher degradation efficiency of azo-reactive dyes which are the most abundant types of dyes found in textile wastewater, has been exhibited by ZnO [14]. In terms of cost, ZnO has been acknowledged as a cheaper alternative to TiO2, which allows its potential to be used in large scale wastewater treatment [15]. In addition to its low cost, ZnO has been reputed for its high stability, wide band gap as well as its high photosensitivity [16]. One of the factors that influence the photocatalytic ability of a semiconductor photocatalyst is its band gap. A larger band gap would require a larger fraction of light to be absorbed in order to facilitate the excitement of photoelectrons from the filled valence band to the empty conduction band. Since TiO2 has a lower band gap compared to ZnO, TiO2 would be expected to have higher photocatalytic abilities than ZnO [7]. However in several studies, ZnO had exhibited photocatalytic abilities surpassing that of TiO2 [17]. This may be due to ZnO intrinsic defects of having positively charged Zn interstitials and oxygen vacancies which help trap photo-electrons. The trapping of photoelectrons help facilitate redox reactions and prevent recombination of electron-hole pairs that are crucial for photocatalytic reactions [18]. Another contributing factor for ZnO’s superior photocatalytic abilities is its ability to absorb a wide range of the solar spectrum which affirms ZnO as an ideal semiconductor photocatayst for the degradation of contaminants under solar irradiation [19]. This allows ZnO to be utilised as a more environmentally friendly semiconductor photocatalyst as sunlight is a renewable and readily available energy source.

2.1. Synthesis of NPs

2.1.1 Chemical synthesis of NPs
Chemical-based synthesis methods are the most commonly adopted methods in the synthesis of metal-based NPs. These chemical methods include techniques such as sol-gel, electrochemical and photochemical reduction, polymerization, hydrothermal, sol gel processing, and magnetron sputtering. solvothermal, hydrothermal, chemical precipitation and simple solution-based synthesis techniques [20,21,22]. However, most chemical methods for the synthesis of NPs involve expensive materials, complicated equipment and the production of toxic by-products [23]. These toxic by-products would get adsorbed onto the surface of the NPs, rendering them unsafe for medical purposes Due to the adverse effects of chemical synthesis methods and the need for more sustainable and environmentally-
friendly processes, green synthesis of NPs has been extensively investigated and studied for its potential in replacing chemical methods as a more sustainable alternative [24].

2.1.2 Green synthesis of NPs
Green synthesis has been developed for decades as an alternative method that utilizes natural plant extracts as a reducing agent instead of harmful chemicals for the reaction. This method is not only a more eco-friendly method of synthesising photocatalytic NPs but is also more economically efficient in comparison to its more conventional counterparts [11,25]. Green synthesis, also known as biosynthesis, is a method of synthesising NPs by the use of plant extracts, enzymes, bacteria and fungi that ultimately produces NPs without the generation of hazardous by-products. In addition, the application of green synthesis over chemical synthesis also aids in reducing energy consumption as chemical procedures often utilise sophisticated equipment and complicated procedures [26,27]. Out of all the different biological sources used for green synthesis, the use of plant extracts offers some advantages over its counterparts such as being easily procured and safe to handle. The process of green synthesis of NPs using plant extracts generally involves the reaction between the plant extract and metal-based precursors. In this reaction, the plant extract acts as a reducing and capping agent that reduces the metal based precursor which acts as an oxidising agent to form metal-based NPs [28,21].

Plant extracts have been produced from different parts of a plant such as its seeds, leaves and flowers and studied for their abilities in synthesising NPs [29,30]. The ability of a plant extract to synthesise NPs lies in the presence of phytochemicals within the plant that are responsible for its synthesising abilities. These phytochemicals include flavonoids, saponins, terpenoids, carbohydrates, alkaloid and protein [31]. Table 1 shows the different types of plant extracts that have been successfully used in previous studies to synthesise various types of metal-based NPs.

| Plant extract          | Parts of Plant Used | NPs produced                | Reference |
|------------------------|---------------------|-----------------------------|-----------|
| Carica papaya          | Seed                | Zinc oxide                  | [30]      |
| Cinnamomum zeylanicum  | Leaf                | Gold                        | [32]      |
| Green tea              | Leaf                | Iron                        | [33]      |
| Ficus benghalensis     | Leaf                | Silver                      | [28]      |
| Carica papaya          | Leaf                | Copper oxide                | [34]      |
| Hibiscus rosa sinensis | Leaf                | Gold and silver             | [35]      |
| Coffee and Tea         | NA                  | Silver and palladium        | [36]      |
| Sargassum muticum      | Seed                | Iron oxide                  | [37]      |
| Jatropha curcas        | Seed                | Silver                      | [38]      |
| Argemone maxicana      | Leaf                | Silver                      | [39]      |
| Nyctanthes             | Leaf                | Titanium dioxide            | [40]      |
| Ocimum sanctum         | Leaf                | Silver                      | [41]      |
2.1.3 Green synthesis of ZnO
ZnO has been acknowledged as one of the most useful metal oxides to be utilised in the nanoparticle form because of its distinctive electrical and optical properties [42]. Due to these properties, ZnO NPs have displayed useful characteristics which have allowed them to contribute to the development of various industries such as the pharmaceutical industry, cosmetics industry and the wastewater treatment industry. In addition, ZnO NPs are also used in the making of light-emitting devices, solar cells and gas sensors [43]. ZnO NPs are extensively used in the pharmaceutical industry due to its antimicrobial, antifungal and anticancer properties [26,27]. In the cosmetics industry, ZnO NPs have been employed in the production of sunscreens and cosmetics due to its ability to efficiently absorb UV-A and UV-B light that would have damaging effects on the skin if not filtered [27]. It is a known fact that ZnO in general is an especially efficient semiconductor photocatalyst that can be effectively utilised under both UV and visible light irradiation. Moreover, ZnO when in the form of NPs have displayed improved photocatalytic abilities especially in the photocatalytic degradation of dyes and organic contaminants in water bodies [23,24,26]. This is due to the larger surface area of ZnO NPs compared to its bulk form as well as its ability to confine charge carriers that are pivotal in the degradation of pollutants under irradiation. Table 2 displays the different types of plant extracts used in past studies to synthesise ZnO NPs. It may be concluded that zinc nitrate is commonly used as the precursor agent for the biosynthesis of ZnO NPs.

| Plant extract            | Precursor                                      | Reference |
|--------------------------|------------------------------------------------|-----------|
| Aloe vera leaf extract   | Zinc nitrate, Zn(NO$_3$)$_2$                  | [24]      |
| Cassia fistula leaf extract | Zinc nitrate hexahydrate, Zn(NO$_3$)$_2$.6H$_2$O | [23]      |
| S. nigrum leaf extract   | Zinc nitrate, Zn(NO3)                         | [27]      |
| Moringa oleifera leaf extract | Zinc nitrate hexahydrate, Zn(NO$_3$)$_2$.6H$_2$O | [26]      |
| Aspalathus linearis flower extract | Zinc nitrate, Zn(NO$_3$)$_2$.6H$_2$O | [29]      |
| Carica papaya seed extract | Zinc gluconate hydrate, C$_{12}$H$_{22}$O$_{14}$Zn.xH$_2$O | [30]      |
| Corriandrum sativum leaf extract | Zinc acetate dehydrate, Zn(O$_2$CCH$_3$)$_2$(H$_2$O)$_2$ | [31]      |

2.1.4 The use of Coriandrum sativum leaf extract to synthesize ZnO
In this example we can see that there are footnotes after each author name and only 5 addresses; the 6th footnote might say, for example, ‘Author to whom any correspondence should be addressed.’ In addition, acknowledgment of grants or funding, temporary addresses etc might also be indicated by footnotes. Coriander (Coriandrum sativum L.), is a plant of the Umbelliferae (Apiaceae) family. It is a native species for the Mediterranean region and widely grown in Asia, Russia, Central Europe, North Africa and South America [44, 45]. It grows to about 20–70 cm in height with green lanceolate-shaped leaves, white or pink umbellate flowers and pronounced taproot. The seeds are globular dry schizocarp with multiple longitudinal ridged surface [46]. The C. sativum seed contains triglyceride oil, petroselinic acid and monounsaturated fatty acid. Thus, the plant is a potential source of lipids. Furthermore, coriander leaves contain anthocyanins that can be biosynthesized or enhanced by salicylic acid and micro elements, especially zinc application [47].

Coriander has been used since ancient times for cooking, medication and flavouring. Among the medication properties, coriander extracts has been reported to exhibit wide range of biological activities such as anticancer, antioxidant and anti-diabetic properties. Other medicinal properties
include anti-mutagenic, anthelmintic, sedative-hypnotic, diuretic, antifungal, hepatoprotective, anti-protozoal, anti-ulcer, post-coital, antifertility and cholesterol lowering. Lastly, it is reputedly protective against lead toxicity and heavy metal detoxification along with neuroprotective, anxiolytic, hypnotic, anticonvulsant, analgesic, anti-inflammatory and antidiabetic [46,48]. Its essential oil has biologically active components rich in geranyl acetate, linalool, nerol, α-terpinene, bornel, α-pinene, cymene, linalyl acetate and gereniol [49]. Coriander seeds and leaves are increasingly used as condiment in food industry, to add flavour to wide range of commercial foods such as meat products, pickles, liqueurs and teas. In addition, the seeds have their traditional medicinal properties and ailments such as spasm, neuralgia, gastric complaints, dysentery, dyspepsia and giddiness [50].

| Table 3. Phytochemical analyses of Coriandrum sativum extract [31] |
|---|---|---|---|
| No | Phytoconstituents | Reagents | Aqueous |
| 1 | Alkaloids | Mayer’s | + |
| | | Wagner’s | + |
| 2 | Carbohydrates | Molisch’s | + |
| | | Benedict’s | + |
| 3 | Glycosides | Legal’s | + |
| | | Borntrager’s | + |
| 4 | Steroid | Libermann burchard’s | + |
| 5 | Fixed oils | Spot test | + |
| 6 | Saponins | Gelatin | - |
| | | Lead acetate | - |
| 7 | Tannins | Ferric chloride | + |
| | | Wagner’s | + |
| 8 | Protein | Millon’s | + |
| | | Biuret | + |
| 9 | Flavonoids | Alkaline Reagent | + |
| | | Shinoda’s | + |
| 10 | Terpenoids | Thionyl chloride | + |

+ Presence - Absence

Coriandrum sativum contains phytochemicals that are responsible for its antioxidant or reducing properties which are flavonoids, saponins, terpenoids, carbohydrates, alkaloid and protein. In addition to its antioxidant properties that aid in the reduction of metal-based precursors to produce NPs, Coriandrum sativum also makes an ideal plant to be used for green synthesis as it can be easily obtained from local supermarkets or grocery stores and is a relatively cheap source of plant extract. Table 4 below shows the type of NPs that have been produced using Coriandrum sativum.
Table 4. Review of Coriandrum sativum as synthesis for NPs research

| No | Type of NPs | Part of Coriandrum sativum Used | Particle Size (nm) | Reference |
|----|-------------|---------------------------------|-------------------|-----------|
| 1  | Silver      | Leaf                            | 6.45              | [51]      |
| 2  | Zinc Oxide  | Leaves and seeds                | 552.3±61.1        | [52]      |
| 3  | Zinc Oxide  | Leaves                          | 60                | [53]      |
| 4  | Iron oxide  | Leaves                          | 20-90             | [54]      |
| 5  | Silver      | Leaves                          | 37                | [55]      |
| 6  | Zinc Oxide  | Leaves                          | 52-253            | [56]      |
| 7  | Silver      | Seed                            | 13.09             | [57]      |
| 8  | Zinc Oxide  | Leaves                          | 66                | [31]      |
| 9  | Silver      | Leaves                          | 26                | [58]      |
| 10 | Gold        | Leaves                          | 6.75–57.91        | [59]      |

2.1.5 Photocatalytic degradation of dyes

In pursuit of discovering new technologies to effectively remove organic and inorganic contaminants in water sources that are highly resistant to traditional treatment methods, photocatalytic degradation using semiconductor photocatalysts has been developed and proven to be a more effective mechanism. In comparison to conventional treatment technologies, photocatalytic degradation have shown several advantages such as the ability to completely degrade and oxidise contaminants into CO$_2$ and H$_2$O as well as the ability to break down highly stable compounds which traditional processes would not be able to achieve [60]. By using a suitable semiconductor photocatalyst in the presence of light, photocatalysis have shown to accelerate the deterioration of a wide range of environmental and highly toxic contaminants [61]. In addition, the cost of operation for photocatalytic degradation is relatively lower compared to other processes which sometimes require disposal of sludge. In selecting the ultimate semiconductor catalyst, several requirements should be met such as activation when exposed to UV, visible and solar light, chemical and biological durability, stability against photocorrosion, non-toxicity as well as being affordable and easily obtained [62]. Many studies have been previously conducted to evaluate and analyse the efficiency of various types of semiconductor photocatalysts in the removal of commercial dyes commonly used in the textile industry. Some examples of these semiconductor photocatalysts are titanium dioxide (TiO$_2$), zinc dioxide (ZnO), tungsten trioxide (WO$_3$), cerium dioxide (CeO$_2$) and tin dioxide (SnO$_2$). Out of these, TiO$_2$ and ZnO are two of the most widely investigated semiconductor photocatalysts for the purpose of identifying the ideal semiconductor photocatalyst to be used commercially in water treatment, specifically for coloured wastewater. TiO$_2$ is regarded as a photocatalyst that has considerable stability towards light and chemicals, relatively high efficiency in the degradation of dyes and is generally non-toxic. However, due to its relatively high cost, the employment of TiO$_2$ for extensive use in water treatment is considered to uneconomical. This has prompted the search for a cheaper semiconductor photocatalyst alternative [7]. ZnO is a similar widely known semiconductor photocatalyst and is relatively cheap in comparison to its counterparts and has the potential to be employed for large-scale textile wastewater treatment. In addition to its economic advantages, ZnO has shown superior photocatalytic activity in comparison to TiO$_2$ in many incidents. [7] reported that the efficiency of degradation of three commercial dyes which were Crystal Violet (CV), Basic Blue (BB) and Methyl Red (MR) without the presence of light for a degradation period of 3 hours. The degradation efficiencies of these semiconductor photocatalysts were then compared to that of Degussa P-25. It was found that ZnO displayed the highest percentage of decolourisation of CV and BB in comparison to TiO$_2$, SnO$_2$ and Degussa P-25, while the decolourisation of MR was negligible for all the semiconductor photocatalysts.
3. Conclusion
The outcome of this study is intended to provide a better solution to water treatment agencies specifically in the area of coloured wastewater treatment. Traditional water treatment methods are not ideal when it involves dyes and the use of photocatalytic NPs will help fill in that gap. The use of ZnO NPs in particular will benefit the water treatment industry in terms of economical values and efficiency in treating coloured wastewater. Furthermore, adopting green synthesis in place of traditional methods of synthesising NPs will allow water treatment organizations to produce photocatalytic NPs without producing toxic by-products. Not only will it serve as a non-toxic alternative, it will also aid in reducing energy consumption by utilizing renewable solar energy. As the current world is moving towards sustainability and conserving the environment being more critical than ever, it is pivotal that we discover more eco-friendly methods of procedure in any given field to reduce their negative implications on the environment. In addition, this study will also contribute to the field of education and scientific research. The outcome of this study will hopefully serve as a guide to anyone intending to conduct research on a similar field of interest. Moreover, this route is greatly applicable for biomedical, cosmetic, gas sensing and agricultural applications due to their non-toxicity. Further exploration on this subject will allow the improvement and evolution of water treatment methods towards a more sustainable approach.

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4. References
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