Research Article

Synthesis of Gold Nanoparticles Using Tannin-Rich Extract and Coating onto Cotton Textiles for Catalytic Degradation of Congo Red

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Gold nanoparticles (AuNPs) were synthesized under ambient conditions from chlorauric acid in aqueous solution at pH 4. Tannin-rich extract from Xylocarpus granatum bark was used as both reducing and capping agent, rapidly converting Au(I) salt to AuNPs. Transmission electron microscopy showed the as-prepared AuNPs to be predominantly spherical, with an average diameter of 17 nm. The AuNPs were tested for catalytic reduction of Congo red (CR), a carcinogenic azo dye, in aqueous sodium borohydride solution. Cotton samples were coated with the AuNPs, taking on a reddish-purple color. The samples showed significantly reduced tearing strength after coating, though tensile strength was unaffected. UV-visible spectroscopy was used to determine the dye concentration in the water. CR degradation was observed only when AuNPs were present, and the efficiency of degradation was strongly linked to the AuNP loading. The AuNP-coated fabrics left only a 4.7% CR concentration in the solution after 24h and therefore promise as a heterogeneous catalyst for degradation of CR in aqueous solution.

1. Introduction

Nanostructured materials are known to be promising candidates for conversion or removal of toxic pollutants. Nanosorbents, nanoparticles, and nanocatalytic membrane systems have demonstrated effectiveness in low-energy treatment of wastewater [1]. Gold nanoparticles (AuNPs) offer a large surface-to-volume ratio, low toxicity, good biocompatibility, and unique optoelectronic properties [2]. They are increasingly used as sensory probes as well as in drug delivery, catalysis, and electronic devices [3–6]. The color of spherical AuNPs depends on their dimensions and may be brown, orange, red, or purple [7]. AuNPs have been used for the catalytic reduction of organic compounds including 4-nitrophenol [8], nitrobenzene, and Congo red [9].

Congo red (CR) is an azo dye. It is carcinogenic and toxic to many organisms. Its use in catalytic degradation has been investigated in many research studies [10, 11]. Naseem et al. [12] present an overview of the use of nanocatalysts for reduction of CR in industrial wastewater. In brief, the reaction between CR and NaBH₄ is thermodynamically possible but not kinetically practical, given the very low reaction rate. Interaction between borohydride ions and CR molecules requires a catalyst surface to which they can attach. The large surface area of nanocatalysts makes them suitable as an electron transfer mediator, promoting electron transfer from the borohydride ions (electron donor) to dye molecules (electron acceptor). If the correct metal nanoparticles are used, the reduction process then becomes kinetically feasible. A range of nanocatalysts have been employed in CR reduction, and rapid catalytic degradation of the dye has been demonstrated. To avoid overlapping between the surface plasmon resonance band of the nanoparticle and the absorption band of the dye, very small amounts of metal
nanoparticles must be used. During reduction, the azo bonds of CR are cleaved to form aromatic amines [12]. Wastewater degradation of CR by AuNPs with sodium borohydride (NaBH₄) has also been demonstrated [13]. Cleavage at the azo bond releases products that have lower toxicity and are less environmentally damaging [12]. Figure 1 shows the decomposition products [13].

Nanomaterials can be derived from biological sources, including nanoparticles, wires, flowers, and tubes. Biological entities including plants and microorganisms may act as reducing or stabilizing agents in the formation of nanostructures. The use of organisms in the formation of bio(nano)materials dates back to the 1980 paper of Beveridge et al., [14] who reported that enzymes excreted by Bacillus subtilis are capable of converting metal ions, including Au, to their elemental form, whereas physicochemical methods may require the use of toxic chemicals, whose toxicity may persist in the nanoparticles obtained; biosynthesis is more biocompatible as naturally-occurring compounds are deposited onto the nanoparticle surface [15–17]. This makes the biologically-stabilized nanoparticles more suitable in medical, cosmetic, or food applications [17].

As plant extracts are abundant and easily processed, their use in nanostructure synthesis has attracted considerable attention. Phyto-assisted synthesis is both energy-efficient and cost-effective. It requires no additional reducing agent, surfactant template, organic solvents, or use of hazardous materials [18–21]. However, biosynthesis often produces nonuniform nanoparticles at low yields. This requires fine-tuning of synthesis parameters such as salt concentration, ratio of biological extract to metal salt, pH, temperature, incubation time, and aeration [19, 21].

Many natural products have been used in the synthesis of AuNPs from aurous ion sources including chloroauric acid (HAuCl₄) [22]. The use of plant extracts has been widely reported, including extracts from leaves, fruit, flowers, roots, seeds, and bark [23]. The morphology of the nanoparticles obtained may include spheres, quasispheres, triangles, cubics, pentagons, hexagons, rods, and plates, though spheres have been most frequently reported [20]. Particle size ranges from less than ten nanometers to a few hundred. Salmalia mala-barica gum from Bombax ceiba has served as a reducing and capping agent in the synthesis of AuNPs from chloroauric acid [24]. The authors concluded that hydroxyl groups play a key role in the reduction of aurous ions. The nanoparticles were predominantly spherical with an average size of 12 nm. Bacteria and fungi, as well as their products, have also been used in the synthesis of gold nanoparticles [25].

In this work, AuNPs were produced using bio-assisted synthesis. Extract from Xylocarpus granatum bark was chosen as the reducing agent, as it is known to have good water solubility and low toxicity [26], and is an abundant tannin-rich resource [27]. The tropical tree Xylocarpus granatum is commonly known as the cannonball mangrove. The catalytic reduction of CR in water was carried out both using the as-prepared AuNPs and AuNP-coated cotton. Cotton was selected to facilitate the handling of the nanoparticles and their removal from water after use. The mechanical properties of the coated fabrics were also evaluated.

2. Materials and Methods

2.1. Materials. Chloroauric acid (HAuCl₄), Congo red (C₃₂H₂₂N₆Na₂O₆S₂), sodium borohydride (NaBH₄), hydrochloric acid (HCl), and sodium hydride (NaOH) were of analytical grade and were used without further purification. Whey protein isolate of 90% purity was purchased. Deionized water was used throughout. Tannin was extracted from Xylocarpus granatum by heating the sun-dried bark in water at 80°C for 1 h (water to bark mass ratio of 5:1) followed by spray drying to yield a reddish-brown dye powder with condensed tannin content of 74.0%.

2.2. Tannin-Mediated Synthesis of AuNPs for Direct Use in the Catalytic Reduction of Congo Red (CR). A specified amount of HAuCl₄ was dissolved in DI water at room temperature under vigorous stirring. Tannin powder (0.08 g) was then added. The total solution volume was 500 mL. The pH was adjusted to 4 and the mixture was stirred for a few minutes until the color became reddish-purple. HAuCl₄ concentrations of 0.010 g/L, 0.03 g/L, and 0.050 g/L were used. The colloidal solutions obtained were coded AuNP-1, AuNP-3, and AuNP-5.

2.3. Tannin-Mediated Synthesis of AuNPs and Coating of Cotton Fabrics. Whey protein isolate (WPI) was mixed with DI water at a concentration of 0.2 g/L. Woven cotton samples of 10 cm × 15 cm were coated with the WPI solution using a padding mangle at a nip pressure of 1 kg/cm² to obtain 100% wet pick-up. The coated fabrics were dried at ambient temperature and pressure. The fabrics were coated a second time with colloidal solutions containing AuNPs, prepared using HAuCl₄ at concentrations of 0.1, 0.6, 0.8, and 1.0 g/L. These were padded onto cotton samples, which were coded Cot-01, Cot-06, Cot-08, and Cot-10. The samples were air-dried under ambient conditions.

Excess AuNPs were removed by soaping with 2 g/L AATCC1993 detergent at 60°C for 20 min. Samples were then air-dried.

2.4. Catalytic Reduction of CR. A solution of CR, NaBH₄, and DI water at pH 4 was used in evaluating the catalytic activity of AuNPs. In the first experiment, the as-prepared colloidal solution containing AuNPs was used to directly degrade 0.100 g/L CR. In the second experiment, the AuNP-coated cotton samples were immersed in the 0.020 g/L CR solution. A lower dye concentration was used in the latter experiment because the decomposition efficiency of the AuNP-coated fabrics was significantly lower than that achieved when making direct use of as-prepared nanoparticles.

2.5. Characterization. The morphology of the as-prepared AuNPs was examined using transmission electron microscopy (JEOL, JEM-2100/HR). A scanning electron microscope (JEOL, JSM-5410 LV) was used to study the morphology of the AuNP-coated fabrics. The tensile resistance of the fabrics was tested using a tensile tester (Instron...
5567), following ASTM D5035. Tear resistance was tested using a tear tester (ElmaTear 655), following ASTM D1424. Color values were reported as CIELAB coordinates and measured using spectrophotometry (GretagMacbeth Color i5). The concentrations of the CR solutions were determined using UV-Visible spectrophotometry (PerkinElmer Lambda 25).

3. Results and Discussion

3.1. Synthesis of AuNPs. Successful tannin-assisted synthesis of AuNPs at pH 4 was confirmed as the solution rapidly changed color from yellow to reddish-purple (see Figure 2). This mirrored an earlier report of AuNP synthesis using extract from banana peel at a similar range of pH values (2–5) [28]. This was expected as banana peel also contains phenolic compounds and tannin [29]. Under alkaline conditions (pH > 7), the reaction mixture did not change color within 24 h, suggesting that AuNPs had not formed.

3.2. AuNP Morphology. Transmission electron micrographs (Figure 3) showed the predominant formation of spherical gold nanoparticles with an average diameter of 17 nm, measured using ImageJ software. The reddish-purple color was explained by the heterogeneity of the particles. Due to the range of nanoparticle sizes, the solution comprised red and blue particles, which mixed to yield an overall purple color. The smaller nanoparticles absorbed wavelengths in the blue-green spectrum and reflected red wavelengths. The larger particles absorbed red and reflected blue. The transition from red to blue in gold nanoparticles has been attributed to aggregation [30].

3.3. Morphology of AuNP-Coated Cotton. Figure 4(a) shows scanning electron micrographs of the pristine cotton and Figure 4(b) shows micrographs of Cot-10. The EDX mapping shown in Figure 4(c) confirmed that elemental gold was uniformly distributed across the fiber surface.

3.4. Color Properties of the AuNP-Coated Cotton Fabrics. As the AuNPs were reddish-purple, they imparted this color to the white cotton samples. The color values are reported in Table 1. A stronger fabric color (higher K/S) is associated with a greater AuNP loading. A higher $a^*$ value indicates a redder shade, while a lower $b^*$ value indicates a bluer shade. A lower $L^*$ value indicates a darker shade and was usually associated with a higher K/S value. The color parameters accurately represented the appearance of the coated samples and confirmed they took on a more reddish-purple shade in the presence of an AuNP coating.

3.5. Catalytic Reduction of CR. Both the as-prepared AuNPs and the AuNP-coated cotton fabrics were effective in catalytic reduction of CR in water, using sodium borohydride (NaBH$_4$) as a reducing agent. Figure 5 shows that the CR concentration decreased with contact time, until the curves plateaued. No decomposition of CR was observed under ambient conditions, in the absence of either AuNPs or NaBH$_4$. This confirmed that, in this system, AuNPs were required to catalyze the NaBH$_4$ reaction. A greater AuNP loading was expected to increase dye decomposition. This was confirmed as the CR degradation time decreased from 6 h when using AuNP-3 to only 4 h when using AuNP-5. AuNP-1 and AuNP-3 were added to the CR solutions, yielding residual CR concentrations of 15.9% and 4.6% after 24 h. The AuNP-1 curve took almost 24 h to plateau. As can be seen from Figure 6, the AuNP-coated fabric showed more gradual decomposition of CR than the use of AuNPs directly. In both treatments, a higher AuNP loading on the coated cotton fabrics enhanced CR degradation. CR degradation in the Cot-01 sample was comparable with that of untreated cotton. Over a 24 h period, the concentration of residual dye decreased by 52.4% for Cot-06 and 4.7% for Cot-10. In contrast, increasing the NaBH$_4$ concentration from 1.00 g/L to 2.00 g/L had no significant effect on decomposition (data not shown). This was attributed to the 1.00 g/L NaBH$_4$ concentration being sufficient to completely disintegrate the CR molecules in the system. As the cotton exhibited strong affinity to CR, absorption of CR would add to the catalytic degradation attributable to AuNPs and NaBH$_4$. This is evident from Figure 6, which shows the CR
concentration also decreases mildly with contact time in the sample that was not treated with AuNPs. Partial leaching of AuNPs from the coated fabrics was also observed.

3.6. Mechanical Properties. The WPI-coated fabrics showed significant declines in tearing strength in both warp and weft directions (Table 2). This was attributed to the WPI restricting yarn and fiber movement. This would in turn disrupt stress distribution, lower the tearing strength, and result in early breaking. WPI was necessary to prevent particle agglomeration and increase nanoparticle stability. In a previous study, we prepared silver nanoparticles (AgNPs) in a similar way, using WPI-assisted synthesis with a silver nitrate precursor. Tannin
extract was subsequently introduced to promote adhesion between the WPI-capped AgNPs and cotton samples [26]. Adding AuNPs to the WPI-coated cotton samples produced no further decrease in tearing resistance, with all coated samples showing a reduction in tearing strength of approximately 40% in both warp and weft directions.

The tensile properties of the AuNP-coated fabrics are presented in Table 3. Because the tensile and tearing forces operate in different modes, different effects result from fabric coating. All samples, coated and uncoated, had comparable tensile properties. In contrast with our previous study [31], we observed a marginal decrease in tensile strength after protein coating. However, in this work, the solution had a much lower WPI solid content (0.20 g/L or 0.02% solid content, compared with 1.4% in the previous work). This may have masked any negative effects of WPI on the tensile properties.

### Table 1: Color values and photographic images of AuNP-coated cotton fabrics.

| Sample   | Color value | Fabric image |
|----------|-------------|--------------|
|          | $L^*$ | $a^*$ | $b^*$ | $K/S$ |          |
| Untreated cotton | 95.88 | 7.33 | −27.23 | 0.87 | ![Fabric image](image1.png) |
| Cot-01   | 79.27 | 11.37 | −17.85 | 0.91 | ![Fabric image](image2.png) |
| Cot-10   | 53.91 | 14.70 | −11.80 | 2.21 | ![Fabric image](image3.png) |

### Table 2: Tearing strength values of fabric samples.

| Sample          | Tearing strength (N) |          |          |
|-----------------|----------------------|----------|----------|
|                 | Warp                 | Weft     |          |
| Untreated       | 11.20 ± 0.04         | 10.71 ± 0.04 |          |
| WPI-coated cotton | 7.10 ± 0.04         | 6.64 ± 0.09 |          |
| Cot-01          | 7.09 ± 0.03          | 6.65 ± 0.05 |          |
| Cot-10          | 6.93 ± 0.05          | 6.66 ± 0.06 |          |

### Table 3: Tensile properties of fabric samples.

| Sample          | Tensile strength (N) |          |          |
|-----------------|----------------------|----------|----------|
|                 | Warp                 | Weft     |          |
| Untreated       | 105.85 ± 6.76        | 97.36 ± 5.27 |          |
| WPI-coated cotton | 118.60 ± 6.88      | 109.09 ± 15.71 |          |
| Cot-01          | 123.59 ± 10.65       | 91.17 ± 7.11 |          |
| Cot-10          | 116.11 ± 13.75       | 96.01 ± 9.33 |          |
4. Conclusions

We demonstrated successful synthesis of spherical gold nanoparticles from chloroauric acid and tannin in pH 4 solution under ambient conditions. The particles had a nonuniform size distribution, with an average diameter of 17 nm. The nanoparticles were coated onto cotton samples pretreated with whey protein isolate, via pad-drying. The coating imparted a reddish-purple color to the white fabric. Catalysis of Congo red reduction by both the as-prepared nanoparticles and the nanoparticle-coated samples was demonstrated, though the latter were significantly less efficient. The efficiency of dye decomposition was strongly linked to the nanoparticle concentration. The treatment significantly reduced tearing strength, while tensile strength was unaffected. The successful degradation of Congo red suggests that gold nanoparticle-coated cotton is a promising heterogeneous catalyst for treatment of dye-contaminated water.

Data Availability

The data used to support the finding of this study are included within the article.

Disclosure

This study was presented in part at the 59th Kasetsart University Annual Conference, Bangkok, Thailand (March, 2021).

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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