Green Synthesis Gold and Silver Nanoparticles and Their Potential as Glucose Monitoring on Paper Analytical Device

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Abstract. Controlling the glucose content in the body is crucial to preventing serious diseases. The American Diabetes Association sets the maximum limit for glucose levels in a person's body at 10 mmol/L. In a previous study, the Analytical Paper Device (μPAD) was invented to make glucose detection easy, cheap, and efficient. Unfortunately, the materials used for sample quantification are still not environmentally friendly, and additional enzymes are required. Therefore, in this study, we prepared AgNPs and AuNPs using the green synthesis method by adding a red dragon fruit peel extract mixture in the experimental process. To ensure the formation of AuNPs and AgNPs was successful, we used a laser to observe a clear strike line, indicating the nanoparticles' presence in the dispersion. The absorbance peaks were observed using UV-Vis spectrometry; the formation of AgNPs was detected within the range of 400 nm, while AuNPs were formed in the 500-600 nm range. We also measured the particle size using PSA. Immobilization is done by dripping the sample on the μPAD and observing its color change after addition. The image analysis was performed using a simple smartphone application, Image-J. Thus, this study set out to develop synthesizing nanoparticles for glucose detection that are naturally degradable and eco-compatible.

Keywords:.

1 Introduction

Among all the biological substances present, glucose is undoubtedly one of the most essential for sustaining life. As the main energy source for glycolysis and the subsequent processes of aerobic and anaerobic respiration, glucose plays a crucial role in producing significant energy potential necessary for effective growth and reproduction. Starch is formed by condensing glucose from water and carbon dioxide by photosynthesis in plants and cyanobacteria. In food,

ICIESC 2024, September 17, Medan, Indonesia

DOI 10.4108/eai.17-9-2024.2353122

especially those originating from plants, varied carbohydrates have a significant role in determining the texture and taste of food while also acting as a supplementary energy source after intake. Therefore, it is crucial in the first metabolic processes for oxidative phosphorylation and glycolysis to produce glycogens, proteins, and lipids. Therefore, monitoring blood glucose levels is essential in the molecular mechanisms of many complex activities.

At present, plenty of detection techniques have been explored to detect glucose, including copper-iodometric and enzymatic methods such as glucose oxidase (GOx), glucose dehydrogenase, Glucose 1-dehydrogenase, Quinoprotein glucose dehydrogenase, FAD-glucose dehydrogenase, and hexokinase. Meanwhile, despite being effective in detecting glucose, the intricate preparation and unaffordable tools restrict the practical application of these methods. Thus, it is critically important to develop an easy-to-use, quick, affordable, and dependable device for monitoring glucose levels, both for the initial diagnosis of diabetes and for ongoing treatment. (Steiner et al., 2011; Xiong et al., 2015) (Shoji & Freund, 2001; H. Wei & Wang, 2008).

Nanomaterials are particles with sizes in the nanometre range (1-100 nm) that possess properties such as an increased surface area-to-volume ratio. Numerous studies have been accomplished to obtain nanoparticles. Unfortunately, its main ingredients are toxic to the environment. There is a growing trend in research to produce nanoscale particles through different techniques, such as chemical, physical, and green synthesis. Traditional chemical and physical methods often involve high energy consumption, toxic waste, and complex equipment. In contrast, green synthesis utilizes natural materials and microorganisms, offering a more sustainable and environmentally friendly approach. This shift reflects a growing awareness of the negative impacts of traditional methods and innovation to develop more responsible manufacturing processes. Some green materials can also be used as end-capping agents and dispersants simultaneously, reducing energy consumption and avoiding the use of toxic and harmful reagents. In the present day, green synthesis mainly uses microorganisms like fungi, bacteria, and algae or extracts from various plants' leaves, flowers, roots, peelings, fruits, and seeds. Even so, dragon fruit peels, often discarded as waste byproducts, are valuable. They contain phytochemicals like betacyanin, flavonoids, and vitamins, which can be used for various purposes. They are not edible and often get discarded as waste. In this study, we utilized the abundance of phenolic compounds in the dragon fruit peel. Phenolic compounds are known to have antioxidant effects, which help elevate the formulation of nanoparticles. Hence, we proposed a greener way to synthesize nanoparticles by adding dragon fruit extract. Due to their size, shape-dependent optical characteristics, and catalytic solid activity, metallic nanomaterials, particularly gold (Au) and silver (Ag), have been increasingly used in colorimetric analysis in recent years. The colorimetric method is a technique that utilizes color changes to measure the concentration of a substance in a sample. Colorimetric sensing strategies using plasmonic nanomaterials are usually based on a change in the optical properties owing to their aggregation and morphology changes. This method is mainly used in chemical, biochemical, or environmental analysis. (Grasianto et al., 2021, 2023).

Microfluidic paper-based analytical devices (μPADs) have many benefits, such as using cheap, easily accessible, biodegradable, and environmentally friendly paper substrates, not needing to prepare the sample in any special way, and not requiring any special instruments

[36–38]. One of the great features of using colorimetric PADs is the possibility of onsite monitoring with inexpensive portable devices, such as smartphones and laptops, to quantify the color signal from the sensing reaction between the target analyte and the pre-deposited reagents on the porous paper, which can be stored for an extended period. This method's principle is the measurement of absorbance or light transmission at specific wavelengths related to the concentration of analytes in the sample . The theoretical basis of this method is the Beer-Lambert law, which states that absorbance (A) is proportional to concentration (C), light path length (l), and molar absorptivity coefficient (ε). The image of (μPAD) was taken using an iPhone 11 mobile camera with ISO set to 800 and a photo light box to control the light that had been adjusted to 50% brightness.

2 Experimental Section

2.1 Materials

Silver nitrate (AgNO3), Gold (III) chloride (HAuCl4), and Monohydrate Glucose were purchased from Merck Millipore Corp. The preparation of dragon fruit filtrate is described in the next section.

2.2 Example subsection heading

The filtrate of dragon fruit peel was gained by blending 80,1497 grams of chopped dragon fruit peel with 1600 mL aquadest. The mixture was heated at 100°C for 15 minutes. After cooling down, the solution was filtered using coarse filter paper to separate the extract from its solid residue and stored at 4°C to remain stable.

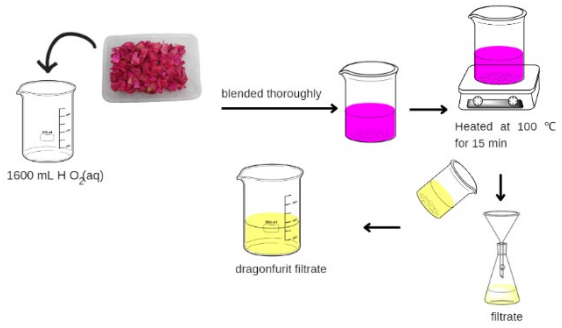

Fig. 1. Preparation of Dragon Fruit Filtrate

2.3 synthesis of nanoparticles

The synthesis of AgNPs was prepared by dissolving 220 mL of the dragon fruit peel extract in 220 mL of 10 mmol L-1 AgNO3. The mixture was stirred for 24 hours in a beaker glass covered with aluminum foil to prevent contamination and indirect light exposure. The solution displays a color change from bright yellow to crimson after being stirred for 24 hours. A similar method was used to synthesize the AuNPs. 220.0 mL of dragon fruit peel extract and 220.0 mL of HAuCl4 solution at 0.25 mmol L-1 were combined. The mixture was stirred for

24 hours in a beaker glass covered by aluminum foil. After the procedure was completed, the solution showed a color change from bright yellow to wine red.

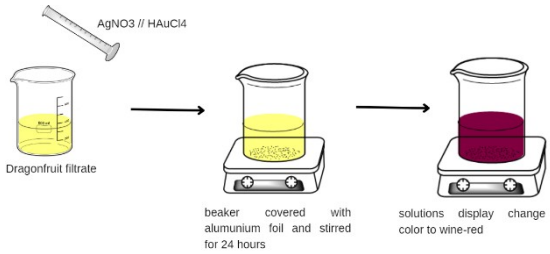

Fig. 2. Synthesis of Nanoparticles

AgNPs and AuNPs were stored at 4°C to keep their stability. After that, the NPs were characterized using laser light, UV-Vis Spectrometer, and Particle Size Analyzer (PSA).

2.4 Fabrication of µPAD

Using the Figma software tool, the paper pattern was created with three vertical and eight horizontal sample zones with diameters of 5.0 mm in every zone and 1.0 mm thick lines inside spaced 1.0 mm apart. The paper sheet size was 100.0 x 50.0 mm (width × length). Next, using the FujiFilm DocuCentre-V C3375 printer, the pattern was printed on filter paper. The printed paper was heated at 165◦C for 60 min and then cooled down to room temperature. The bottom part of the paper is coated with a superabsorbent material to prevent leaking. The procedure for Glucose detection using the µPAD is described as follows, 100 µL of AgNPs or AuNPs solution was dropped into the hydrophilic zones of the PAD. After that, 100 µL of the glucose solutions at various concentrations ranging from 0.50 µM to 200.00 µM were added onto the top of the nanoparticle solution on the detection zones. After the solutions had completely dried and displayed a color shift, the PAD was captured in a photo light box with the exposure adjusted to 50% using an iPhone 11 smartphone camera with an ISO set to 800.

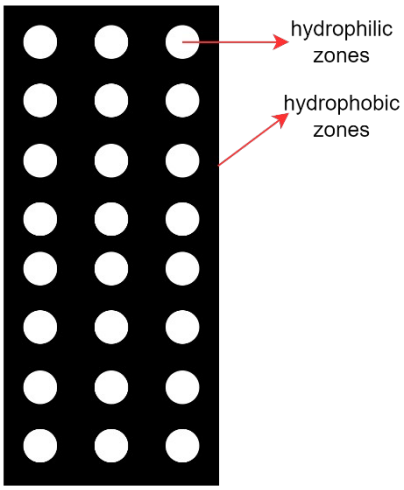

Fig. 3. μPAD Fabrication

3 Result and Discussion

3.1 Result of AgNPs and AuNPs Synthesis

The fundamental idea of the creation of nanomaterial-based glucose sensing is that glucose is oxidized in the presence of oxygen by nanoparticles functioning as a nanozyme, producing gluconic acid and hydrogen peroxide. In the presence of glucose, the color of the hydrophilic zones on the μPAD would change during incubation. This color change is utilized to measure the glucose concentration without relying on any enzyme substrates. According to the UV-Vis and PSA characterization results, it is evident that the optimal NPs were achieved by mixing AgNO3 10 mM and dragon fruit peel filtrate in the 220 mL and 220 mL ratios, respectively. Based on the theoretical framework, AgNPs should be observed around 400 nm. In this assay, we observed a peak within the range of 420 nm, which positively suggests that AgNPs were successfully formed.

3.2 Characterization of AgNPs and AuNPs

Laser light can be used to visually indicate the presence of nanoparticles in a dispersion. If the laser applied to the dispersion forms a clear straight line, we can confirm the possibility of nanoparticles being present in the dispersion. So, we observed the NP dispersion right after. The result shows a clear, straight line between AgNPs and AuNPs, as shown in Fig 4 and Fig 5.

Fig. 4. AgNPs Dispersion

Fig. 5. AuNPs Dispersion

The absorption spectrum of both nanomaterials in the UV-Vis spectra showed two dominant absorption bands ranging from 400 nm and 500 nm for AgNPs and AuNPs, respectively, as shown in Fig. 8. Correspondingly, as shown in Fig 5., The Particle Size Analyzer (PSA) confirms that the particle size of both AgNPs and AuNPs fits within the nanomaterials range. AgNPs have an average size of 161 nm, whereas AuNPs have an average size of 127 nm.

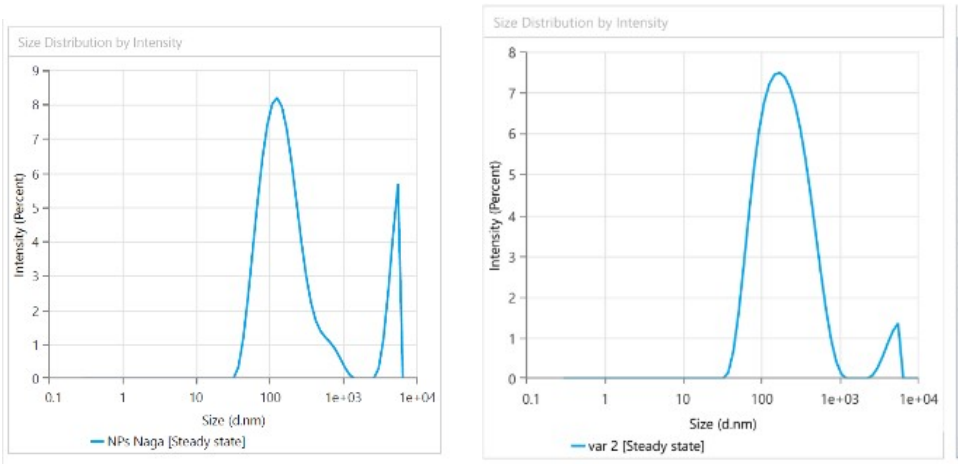

Fig. 6. Size distribution profile (i) AuNPS and (ii) AgNPS

By linear graph analysis, we examined the correlation between lightness (ΔL) and glucose concentration within the range of 0 mg/dL to 200 mg/dL. The data in Fig 7. demonstrates a clear inverse correlation, suggesting that higher concentrations displayed a more intense appearance. This sensor is capable of detecting glucose at a higher concentration, as evidenced by its R2 value of 0.9793. These experiments proved the capability of our designed sensor to detect glucose at elevated concentrations.

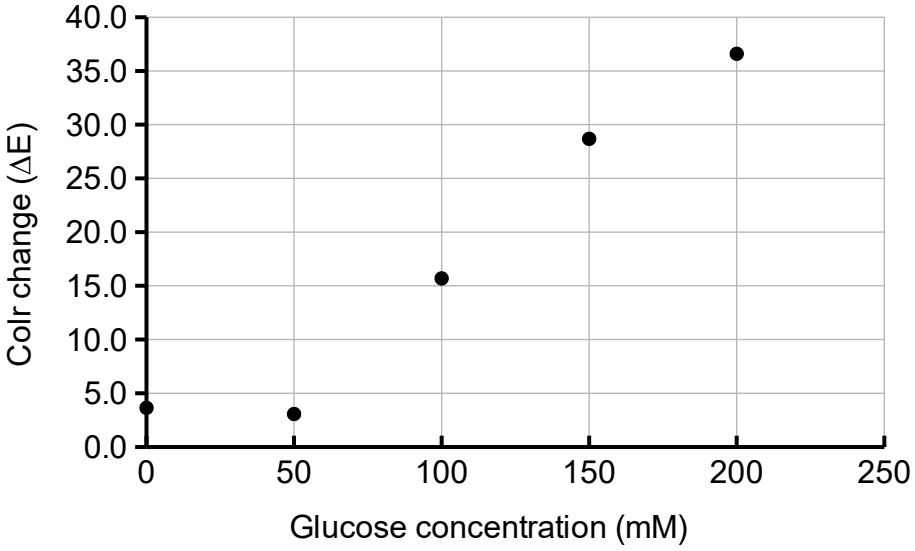

Fig. 7. Color change response (ΔE) to various concentrations of glucose

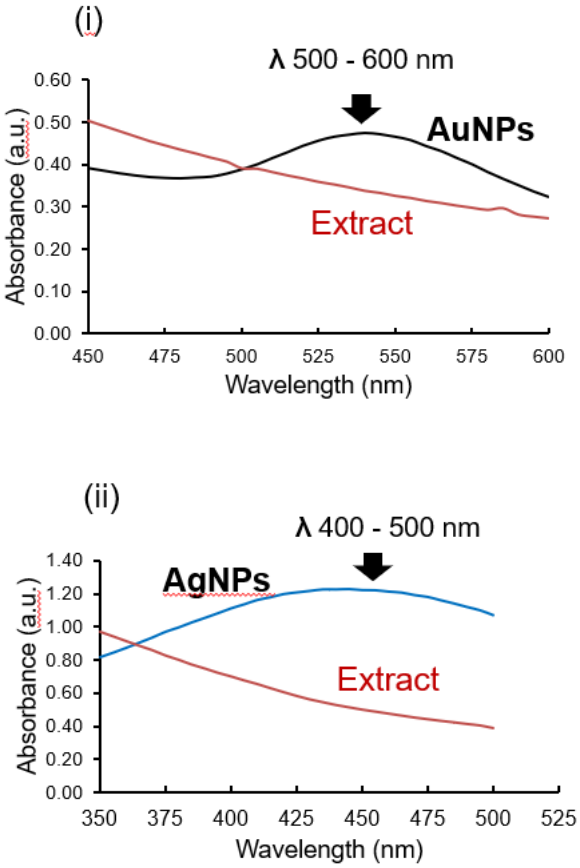

Fig. 8. UV spectra of AuNPs (i) dan AgNPs (ii)

4 Conclusion

This study presents an innovative method to enhance stability and cost-effectiveness in glucose monitoring, utilizing nanomaterials derived from the green synthesis material dragon fruit peel filtrate. The nanomaterials consist of AuNPs and AgNPs immobilized in basic µPADs. This paper-based sensor has been proven to detect a wide range of glucose concentrations, showing its ability to detect low or high doses of glucose from the sample. Although the current study is based on an artificial sample, the findings suggest that the dragon fruit peel extract is promising for green-synthesizing nanoparticles.

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