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Green synthesis of silver nanoparticles and designing a new amperometric biosensor to determine glucose levels

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ABSTRACT

The high-level of glucose in daily nutrition causes diabetes (Diabetes mellitus) and obesity. It is also important to determine glucose levels in food production processes for quality control methods. Biosensors are bio analytical devices that provides cheap, simple, analyses and have a fast response time. Silver nanoparticles are used to modify electrodes in bio-sensing glucose. The nanoparticles could be synthesized via the green synthesis technique. In this technique, plants could be used both reducing and stabilizing agents. Agricultural waste, an environmental problem, is increasing with the population. These wastes are a good natural source and have the potential for green synthesis of silver nanoparticles to use in biosensor applications. In this study, a new amperometric glucose biosensor was designed. For this purpose, carbon paste electrode (CPE) was modified by green synthesized waste tea based silver nanoparticles (WT-AgNPs) and the glucose oxidase enzyme was immobilized on MCPE by the cross-linking. Glucose determination was performed based on the reduction of hydrogen peroxide at +0.4 V versus Ag/AgCl. The linear working range was determined as 0.10–1.0 μM. In addition, the designed biosensor was applied to detect the amount of glucose in a commercial fruit juice. The results showed that the designed biosensor has low detection limit, very good reproducibility, selectivity, and almost long shelf life.

1. Introduction

Glucose has an important role in many metabolic reactions. It plays an active role in producing the body's energy and is converted into metabolic intermediates necessary for the maintenance and division of body tissues. The amount of glucose that the body needs per day is 35 g (150 calories) for women and 20 g (100 calories) for men (Stanhope et al., 2011). The determination of lower glucose levels in blood has importance in the early diagnosis and treatment of many diseases. The high-level of glucose in the blood could be related to the Diabetes mellitus and daily nutrition (Panagiotakos et al., 2005). Excessively consumed fruit juices raised the blood glucose level and cause diabetes (Diabetes mellitus) and obesity (Murphy et al., 2017). According to the studies carried out by the World Health Organization, while 171 million people in the world have diabetes in 2020, they have predicted that this number will approach 366 million in 2030 (Saeedi et al., 2019). In addition the health conditions, it is also important to determine glucose levels in food production processes for quality control methods. Sugars, alcohols, phenols, oligonucleotides, and O_2 need to be detected at many levels of the manufacturing process, as well as in the final product (Artigues et al., 2017). Thus, glucose levels must be determined rapidly, cheaply, and economically both in blood and food samples even if lower concentrations. Different techniques such as chromatography, and spectroscopy are used to determine glucose level but they are expensive, and required well experienced personnel. In addition, interfering agents such as ascorbic acid could be separated by using different techniques such as solid phase extraction (SPE) or chemical modifications. These sample processes increase the complexness, time, and cost of analysis (Artigues et al., 2017).

Biosensors are bio analytical devices that combine with a biological material (enzymes, microorganisms, etc.) and a physical transducer to determine measurable signals of analytes (Karunakaran and Keskin, 2022; Arslan et al., 2022). Biosensors are used in many areas such as medicine, environment, engineering, biology, chemistry, physics, and other industrial applications (Keskin and Arslan, 2020). Biosensors have advantages in determining analytes. They are cheap, simple, and have a fast response time (Arslan et al., 2022; Bodur et al., 2023). To develop glucose biosensor, rapid and accurate determination of blood sugar for

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$$\text{Glucose} + O_2 \xrightarrow{\text{glucose oxidose}} H_2O_2 + \text{gluconic acid}$$

$$H_2O_2 \rightarrow O_2 + 2\,H^{+} + 2e^{-}$$

Fig. 1. Reaction mechanism of H_2O_2 release (Arslan et al., 2012).

the treatment and control of diabetes is important. It is also important for controlling the amount of glucose in food production processes (Arslan et al., 2011).

Nanotechnology is a field of science that examines to obtain nanosized materials and adds innovations in many applications with nanoparticles (1–100 nm) (Parthiban et al., 2019). The interest in applications that use nanoparticles increases because of the features of nanoparticles such as physical, chemical, and biological properties according to their size, distribution, and morphology (Khan et al., 2019). Thus, they could be used in many areas such as health applications, cosmetic products, biomedical devices, mechanics and optics, food and food supplementary, catalysis, drugs, environmental applications, chemical products, electronics and space industries, and energy science (Zhang et al., 2016).

Nanoparticles could be synthesized as organic and inorganic nanoparticles (Li et al., 2022). While carbon nanoparticles (quantum dots, fullerenes, nanospecies) are organic nanoparticles; semiconductor nanoparticles (ZnS, CdS, ZnO), metallic nanoparticles (Ag, Au, Al, Cu) and magnetic nanoparticles (Co, Ni, Fe) are inorganic nanoparticles (Rafique et al., 2017). Silver nanoparticles (AgNPs) have a significant surface area leading to remarkable biochemical reactivates, catalytic activities, and atomic behaviors associated with larger particles with the similar chemical structure and thus have a wide range of applications (Maier et al., 2003; Xu et al., 2006; Khan et al., 2019). In general, nanoparticles could be synthesized in chemical and physical techniques which are quite expensive and potentially dangerous for the environment, using of toxic and hazardous chemicals responsible for various biological risks (Ahmed et al., 2016). Because of these disadvantages, the environment-friendly green synthesis technique was developed (Singh et al., 2018). The green synthesis method has advantages such as being environmentally friendly, cost-effective, nontoxic solvents are used, the process being biocompatible, and higher stability (Thakkar et al., 2010). Plants, fungi, and algae are generally used as reducing agents in green synthesis (Rafique et al., 2017).

Agricultural and herbal household food wastes are increasing day by day due to the increasing population. The average amount of herbal household food waste per person in the world is 0.74 kg (Abdullah et al., 2022). The prevention or recovery of waste is extremely important due to the high amount and rate of increase. Agricultural wastes with low inorganic and high carbon content and bioactive compounds could be used a precursor for production nano materials. Many agricultural wastes such as olive, apricot, cherry and stones, cotton stalks, sunflower shells, etc. could be used in different applications of nanotechnology (Nguyen et al., 2019). Black tea, which is the second most consumed beverage in the world after water, is produced from the production of the *Camellia sinensis* L. plant. (Vinson and Dabbagh, 1998). *Camellia sinensis* L. is rich in bioactive compounds such as enzymes, alkaloids, phenolic compounds and carbohydrates, thus waste *Camellia sinensis* L. also contains a certain amount of these bioactive components (Hamilton-Miller, 1995). The amount of waste tea generated is also higher due to excessive consumption (Turgut and Köse, 2015). With the recovery of waste tea active compounds with the re-extraction method, silver nanoparticles could be synthesized, the waste tea could be recycled and used in the field of technology.

In this study, a new amperometric glucose biosensor was prepared to determine fruit juice glucose levels. For this purpose, waste tea leaves were used for green synthesis of silver nanoparticles to obtain an amperometric glucose biosensor. The synthesized silver nanoparticles (WT-AgNPs) were characterized and used to modify the carbon paste electrode to design a new amperometric glucose biosensor. Bovine serum albumin (BSA) and glutaraldehyde (GA) were used to immobilize glucose oxidase on the modified electrode (MCPE) by cross-linking. The optimum pH, temperature, amount of glutaraldehyde, and substrate concentration were identified. In addition, the amount of WT-AgNP, the linear working range of the designed biosensor and the effect of ascorbic acid were investigated. The determination of glucose even at low concentrations with a lower interference effect of ascorbic acid in the complex matrix by using the prepared biosensor was achieved. Thus, recycling the bioactive compounds from herbal household wastes has the potential of use in the field of technology to detect analytes.

2. Material and methods

The electrochemical studies were carried out using an CHI 1230-A electrochemical analyzer with a three electrode cell. The auxiliary and reference electrodes were a Pt wire and an Ag/AgCl electrode (3 M KCl), respectively. A carbon paste electrode with a surface area of 0.6 cm^2 was used as the working electrode. The pH values of the buffer solutions were measured with an HANNA HI-8424 pH/ion meter. Temperature control was achieved with a Grant GD120 thermostat. Glucose oxidase (*Aspergillus niger*, 10KU) and nujol were supplied by Sigma and graphite powder was purchased from Sigma. All other chemicals were obtained from Sigma.

2.1. Preparation and characterization of WT-AgNPs

100 g of tea leaves (*Camellia sinensis* L.) were brewed with 2.0 L distilled water for 20 minutes (classical method) at 80 °C (Salman et al., 2019). After 20 minutes, the tea extract was filtered and the waste tea leaves were obtained. After that, the waste leaves were re-extracted 4 times with 300 mL of distilled water and filtered with Whatman No 1 to obtain bioactive compounds in waste tea leaves. Then, 0.05 M $AgNO_3$ solution was mixed with the obtained extract and stirred at 40 °C for 40 min (Keskin et al., 2023). At the end of the period, the color of the solution was darkened and it was centrifuged at 10.000 rpm for 15 min to obtain waste tea- based silver nanoparticles (WT- AgNPs) (Keskin, 2022).

Obtained nanoparticles were characterized by scanning electron microscopy (SEM) (ZEISS/Supra 40 VP), UV–vis spectrophotometer (Hach, DR/4000 U), Fourier transform infrared spectroscopy (FTIR) (Thermo Fisher), and energy dispersive X-ray (EDX).

2.2. WT-AgNPs modified carbon paste electrode preparation

To prepare the modified carbon paste electrode (MCPE), 0.0650 g of graphite powder, WT-AgNPs (0.00650 g), and 40 μl of Nujol were mixed homogeneously and the electrode chamber was filled with no gap. The surface of the MCPE was cleaned and polished with the special pad (Bodur et al., 2023).

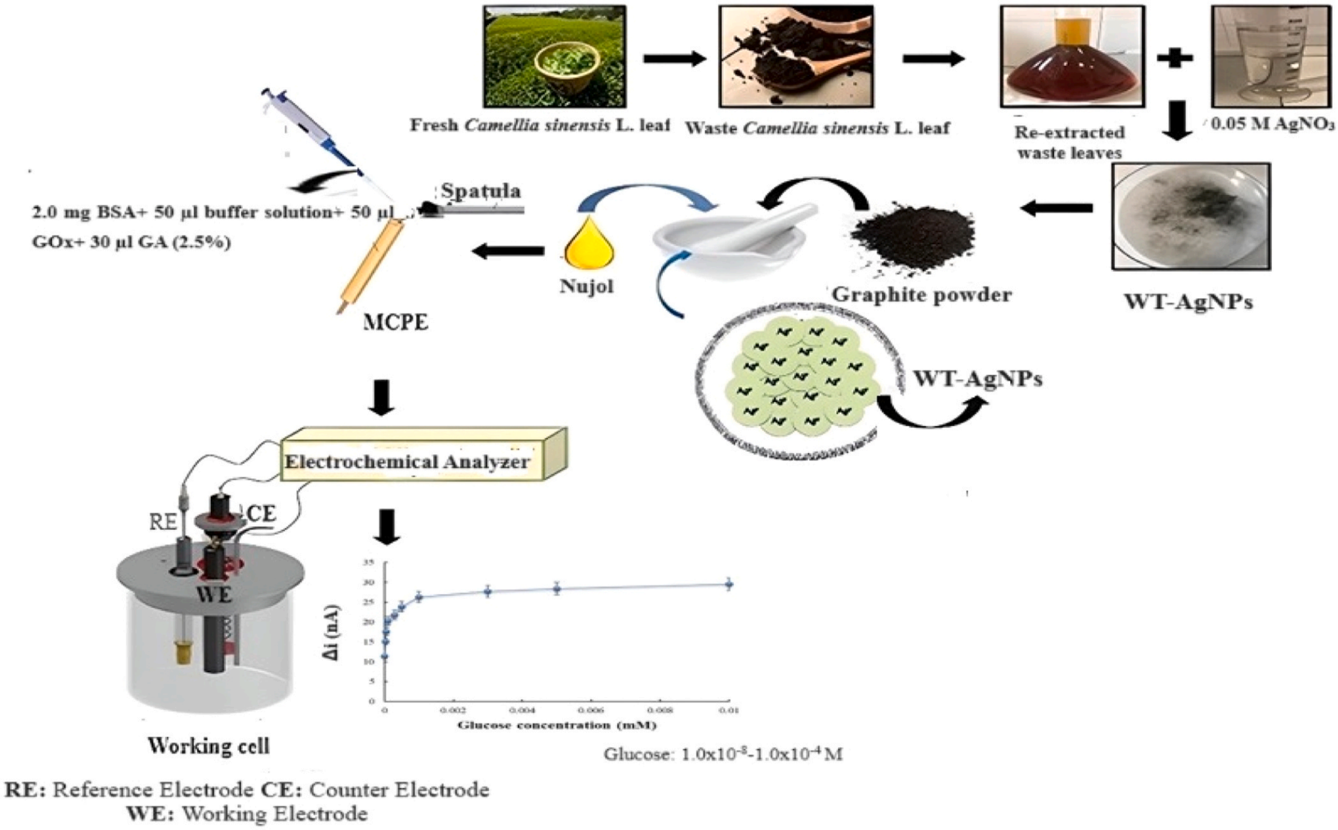


Fig. 2. Green sythesis of WT-AgNPs and electrochemical detection of glucose.

2.3. *Preparation of enzyme electrode and amperometric determination*

To prepare enzyme electrode, 2.0 mg of BSA, 50 µl buffer solution, 50.0 µl glucose oxidase enzyme (1000 Unit mL^{-1}), and 30 µl GA (2.5%) were mixed homogeneously. This mixture was dripped onto the surface of MCPE and dried at room conditions (Arslan et al., 2012). The dried enzyme electrode was cleaned with distilled water to remove the non-crosslinked enzyme from the electrode. The enzyme electrode was kept in phosphate buffer (pH 7.0) at +4°C when not in use.

Glucose detection was based on the oxidation of glucose to gluconic acid and H_2O_2 as a result of enzymatic reaction at +0.40 V versus Ag/AgCl (Fig. 1). It was performed in phosphate buffer solution (pH 7.0), and 0.1 M NaCl as a supporting electrolyte was added to the cell. The MCPE was equilibrated versus Ag/AgCl electrode until obtaining a constant current value. After the equilibrium current (ia) was recorded, glucose solution was added to the cell and the system was stirred. The final current (ib) was recorded at the end of the reaction. The glucose amount was graphed against the current difference. After determination of the optimum working potential, pH, substrate concentration, temperature, and other factors which changes the operation conditions of the biosensor as operational stability, storage stabilization, and interference effects were determined. Finally, the glucose concentration of a commercial fruit juice sample was detected.

3. Results and discussion

A new amperometric glucose biosensor was prepared with the WT-AgNPs modified CPE. For this purpose, WT-AgNPs were synthesized by using waste tea leaves. The enzyme-MCPE was prepared by immobilization of glucose oxidase on MCPE. Determination of glucose concentration can be accomplished through the electrochemical detection of enzymatically released H_2O_2 (Fig. 2). The best operation conditions of the designed glucose biosensor and the key factors involving the performance on the biosensor were investigated.

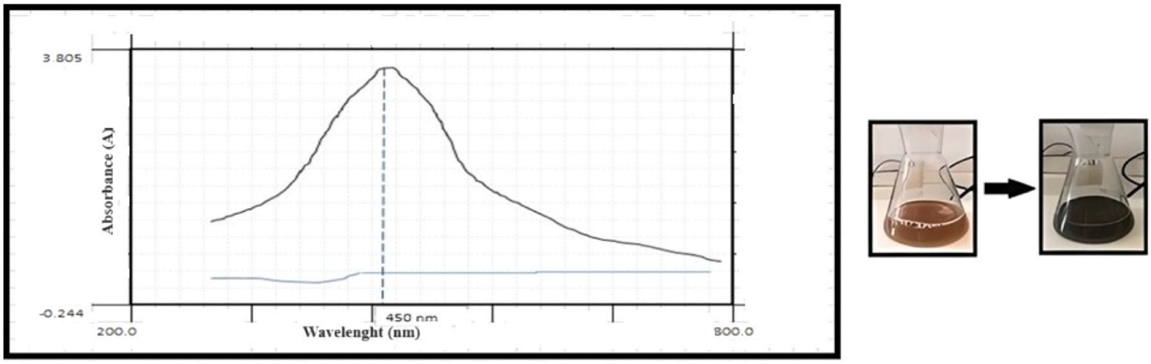


Fig. 3. UV spectrum of a) waste tea leaf extract b) WT-AgNPs.

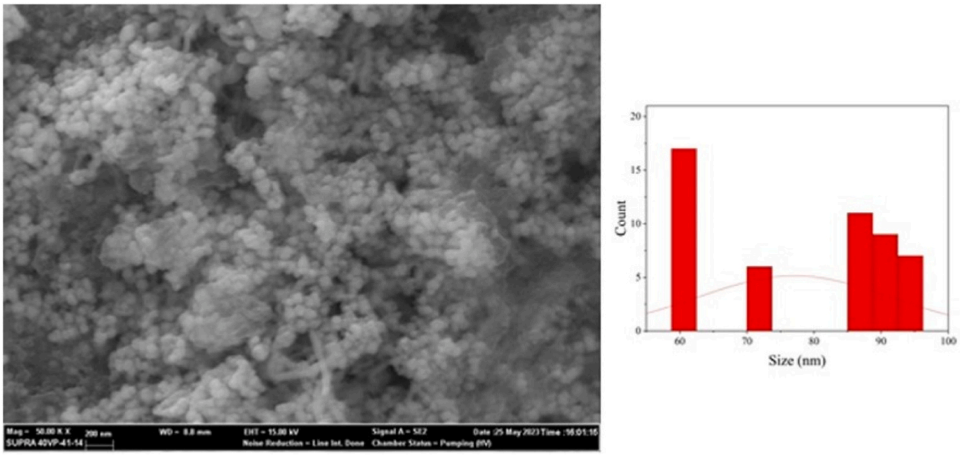
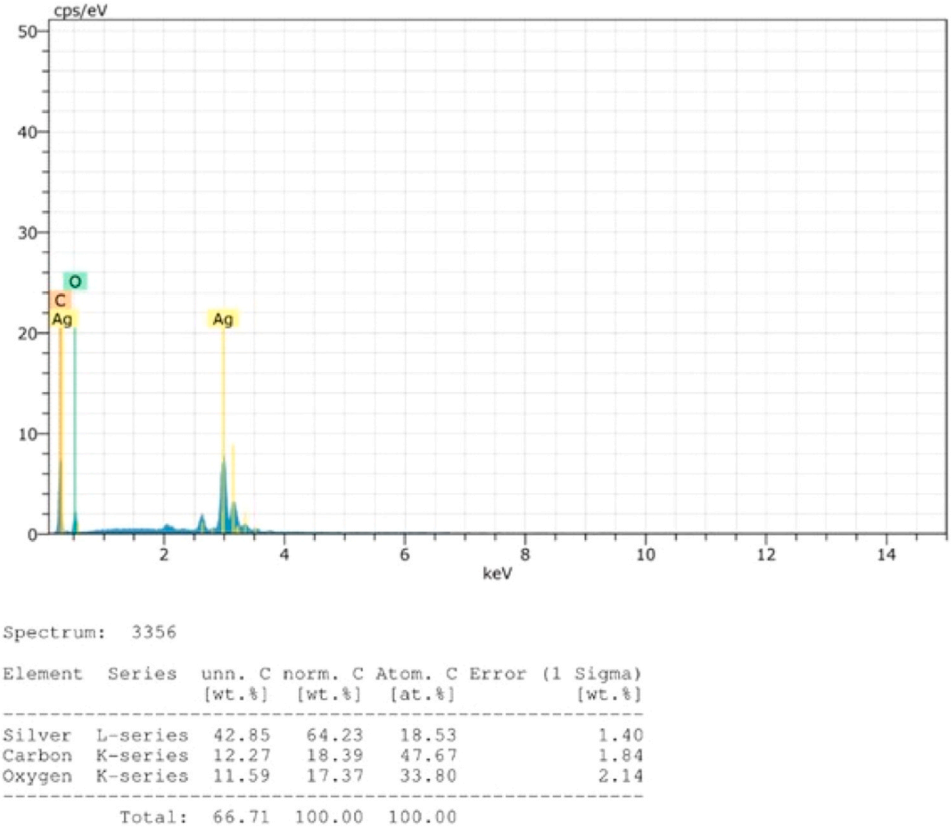


Fig. 4. SEM image and histogram of WT-AgNPs.



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Spectrum:  3356

Element  Series   unn. C norm. C Atom. C Error (1 Sigma)
                  [wt.%]  [wt.%]  [at.%]          [wt.%]
------------------------------------------------------------
Silver  L-series   42.85   64.23   18.53            1.40
Carbon  K-series   12.27   18.39   47.67            1.84
Oxygen  K-series   11.59   17.37   33.80            2.14
------------------------------------------------------------
          Total:   66.71  100.00  100.00
```

Fig. 5. EDX results of WT-AgNPs.

3.1. *Synthesis and characterization of WT-AgNPs*

The synthesized WT-AgNPs were characterized using various techniques to find out potential functional groups, shape, size, and morphology of AgNPs. UV–vis absorption spectroscopy technique was used for characterization of WT-AgNPs and silver nanoparticles showed maximum absorbance at 450 nm (Fig. 3). At the end of the reaction, the solution color changed from yellowish orange to dark brown. This color change is a strong indicator of the formation of silver nanoparticles and is attributed to the adaptation of the surface plasmon of silver nanoparticles (da Silva Ferreira et al., 2017). In this process, ionic silver ion (Ag^+) is reduced to metallic silver (Ag^0) due to bioactive compounds in re-extracted waste tea leaves (Keskin et al., 2023). It was clear that the obtained value is compatible with the literature (Sadeghi and Gholamhoseinpoor, 2015; Anandalakshmi et al., 2016; Liaqat et al., 2022). FTIR, EDX, and SEM were also used for the characterization of WT-AgNPs and particle sizes were found between 59 and 93 nm (Fig. 4) in the SEM. To analyze the dispersed size distribution in the liquid and the main components of NPs, EDX is applied (Strasser et al., 2010; Sankar et al., 2015; Moodley et al., 2018). The presence of Ag was seen with a peak nearly a 3.0 keV in EDX graph (Fig. 5) and it was clear that the carbon (C) and oxygen (O) contents were relatively lower than the elemental silver (normalized atomic value 64.23%). The essential groups which are possibly responsible for the reduction of silver ions to metallic silver were determined by FTIR (Fig. 6). It was determined that there were bands of nearly 3330.81 cm^{-1} for waste tea leaves extract

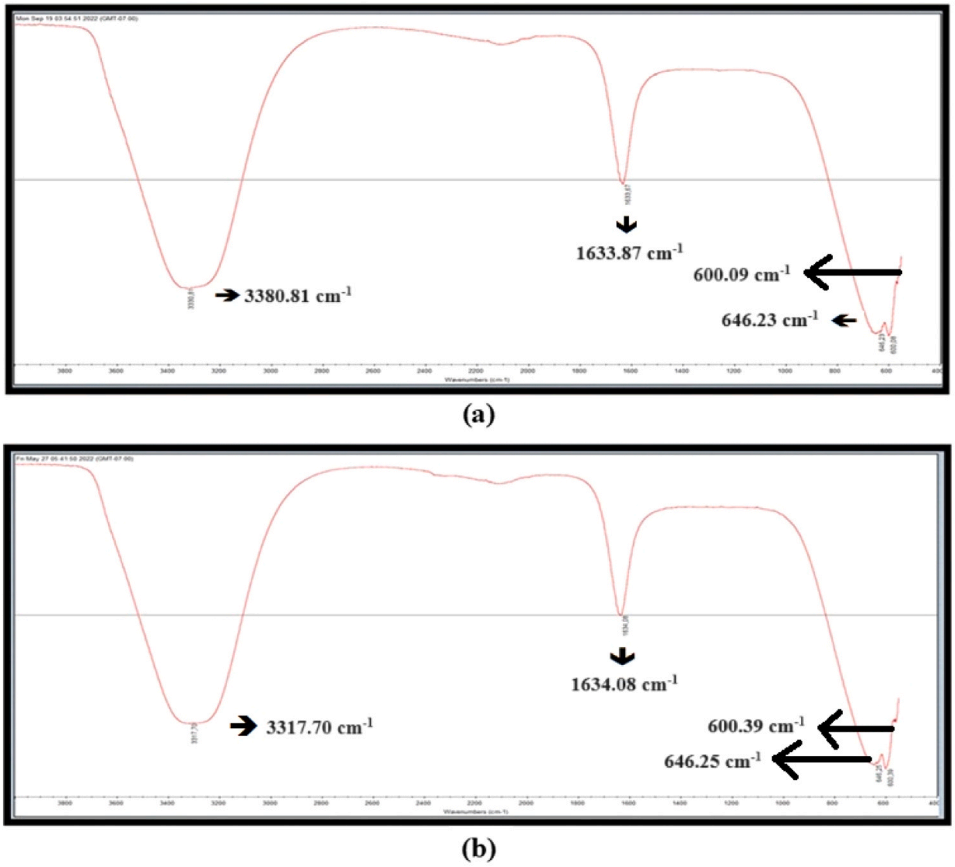

Fig. 6. FTIR spectrum of a) waste tea leaf extract b) supernatant of WT-AgNPs (x axis is wavenumbers (cm^{-1}), y axis is transmittance (%)).

Table 1
The list of FTIR peaks.

	I (cm^{-1})	II (cm^{-1})	III (cm^{-1})	IV (cm^{-1})
Waste tea leaves extract	3380.81	1633.87	646.23	600.09
Supernatant of WT-AgNPs	3317.70	1634.08	646.25	600.39

and 3317.70 cm^{-1} for the WT-AgNPs. These bands in the spectrums indicate the presence of -OH groups. The band at 1633.87 cm^{-1} in waste tea leaves extract and 1634.08 cm^{-1} in WT-AgNP consists of stretching modes depending on the -CO carbonyl group –NH interconnection (Table 1). It also shows strong voting at this wavelength in phenolic and flavonoid images (Jabbar et al., 2020). The peak at 600 nm corresponds to C-Cl stretch selections for alkyl halides (Sadeghi and Gholamhoseinpoor, 2015). The obtained results were compatible with other Ag nanoparticle synthesis processes in the literature (Geoprincy et al., 2013). Small changes occur in the absorption bands between the waste tea leaves extract and the WT-AgNPs spectra, causing a shift of ±1–10 cm^{-1}. According to this comparison, it was clear that the synthesis of nanoparticles with plant extract was produced by some metabolite functional groups such as amines, alcohols, ketones, aldehydes and carboxylic acids (Sadeghi and Gholamhoseinpoor, 2015).

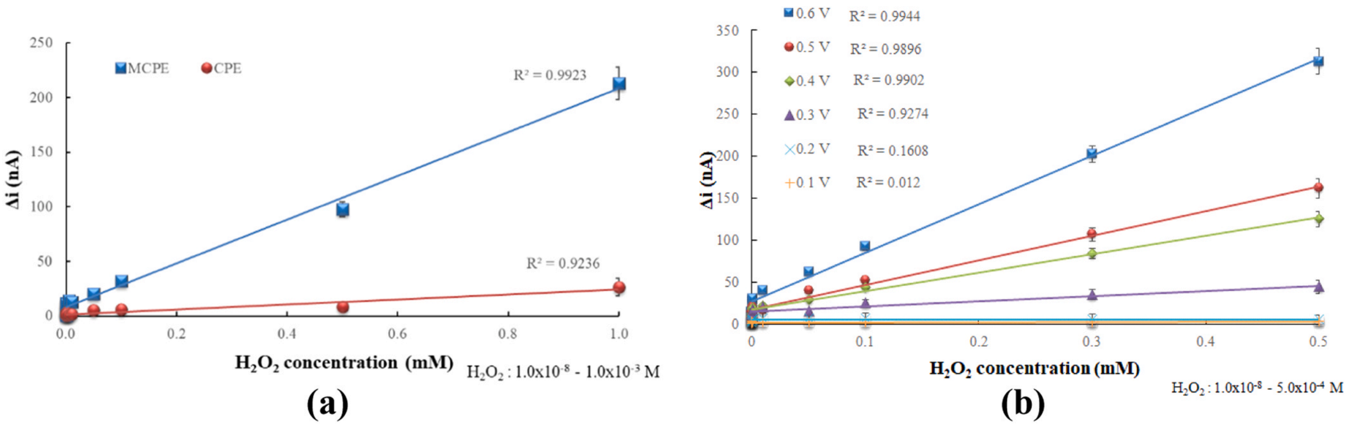

Fig. 7. (a) Comparison of amperometric responses of CPE and MCPE (b) Effect of working potential on H_2O_2 response of MCPE (at 25 °C, in 0.1 M pH 7.0 phosphate buffer).

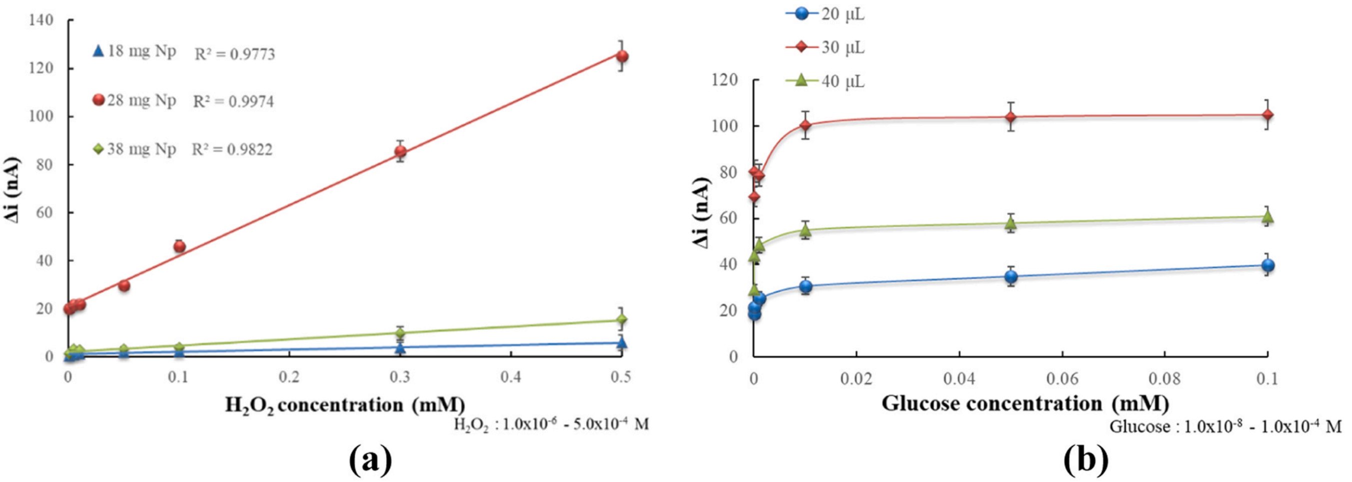

Fig. 8. (a) Effect of WT-AgNPs and (b) Glutaraldehyde amount on amperometric response of MCPE ((+0.4 V operating potential, at 25 °C, in 0.1 M pH 7.0 phosphate buffer).

3.2. *Determination of the response and working potential of CPE and MCPE electrodes to H_2O_2*

The silver nanoparticles could be increased the fundamental analytical properties of biosensors, such as sensitivity, the limit of detection, linear detection range, stability, etc. (Sun et al., 2023). To determine the response of CPE and MCPE, the anodic currents generated by the oxidation of H_2O_2 were compared. For this purpose, CPE and MCPE were equilibrated at +0.4 V. The obtained current differences (Δi) against the H_2O_2 concentration (1.0×10^{-3}- 1.0×10^{-8} M) were plotted (Fig. 7a). It was clear that the obtained currents by MCPE were higher than CPE due to the increasing of conductivity by WT-AgNPs used for modification.

To determine the optimum working potential on the sensitivity of MCPE to H_2O_2, the current differences (Δi) were recorded against the different H_2O_2 concentration (1.0×10^{-3} - 1.0×10^{-8} M) at different potentials (+0.10 V, +0.20 V, +0.30 V, +0.40 V, +0.50 V, and +0.60 V) and graphed (Fig. 7b). It was clear from the graph, the maximum reduction current of H_2O_2 was obtained at +0.60 V, but +0.4 V was used as working potential, which has high response currents, good linearity, high R^2 value and low interference effect. The glucose biosensors studied at different potentials were presented in the literature as +0.4 V (Ren et al., 2005), +0.30 V (Popov et al., 2021), +0.84 (Izadyar et al., 2021) and +0.2 V (Kausaite- Minkstimiene et al., 2020).

3.3. *Effects of WT-AgNPs and glutaraldehyde amounts on the amperometric detection of H_2O_2*

To determine the effect of WT- AgNPs amount on MCPE of H_2O_2 sensitivity, working electrodes with different amounts (18, 28, and 38 mg) of WT- AgNPs were prepared separately. The current differences (Δi) were calculated and graphed against the H_2O_2 concentration for each electrode (Fig. 8a). It was determined that 28 mg of WT- AgNPs showed the best linearity and the highest response current.

Immobilization of enzymes could be performed with different methods as cross-linking, adsorption, covalent linking and entrapment. To use low amount of enzymes for immobilization, cross-linking technique could be used. Glutaraldehyde (GA, $C_5H_8O_2$) is an important crosslinking reagent for the enzyme immobilization. The GA interacts with the amine groups of the enzyme to obtain stable cross-links thermally and chemically (Xiang et al., 2007). Different amounts of GA (10–30 µl (2.5%)) were used to prepare the MCPE to obtain the optimum amount of GA for immobilization of glucose oxidase. The differences of currents were graphed against the glucose concentration (Fig. 8b) and 30 µl GA was determined as the optimum amount of GA.

3.4. *Determination of pH and Temperature effects*

Due to differences of ionizations of the active site or side groups of enzymes, pH is an important parameter that affects the activity of the enzyme. For the best activity, the optimum pH value must be established. To examine the effect of pH on the amperometric response of the

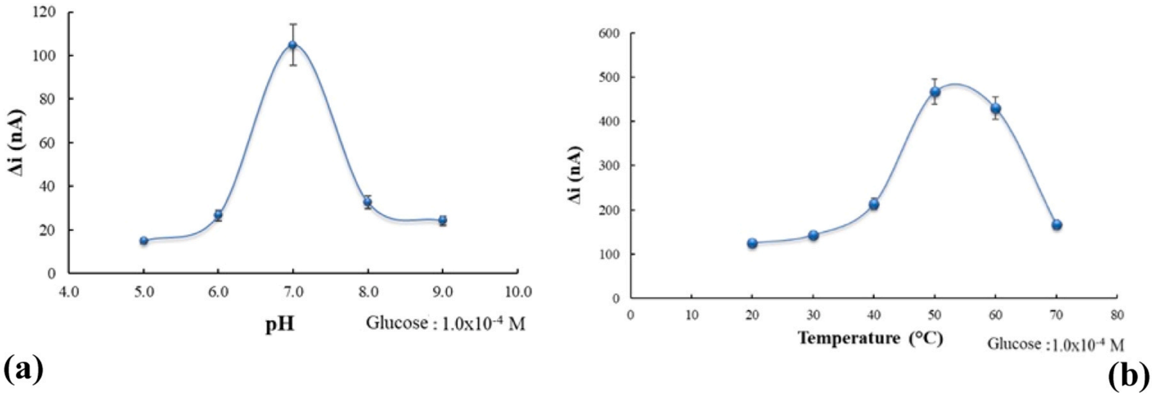

Fig. 9. Effect of (a) pH and (b) temperature on Glucose response of MCPE.

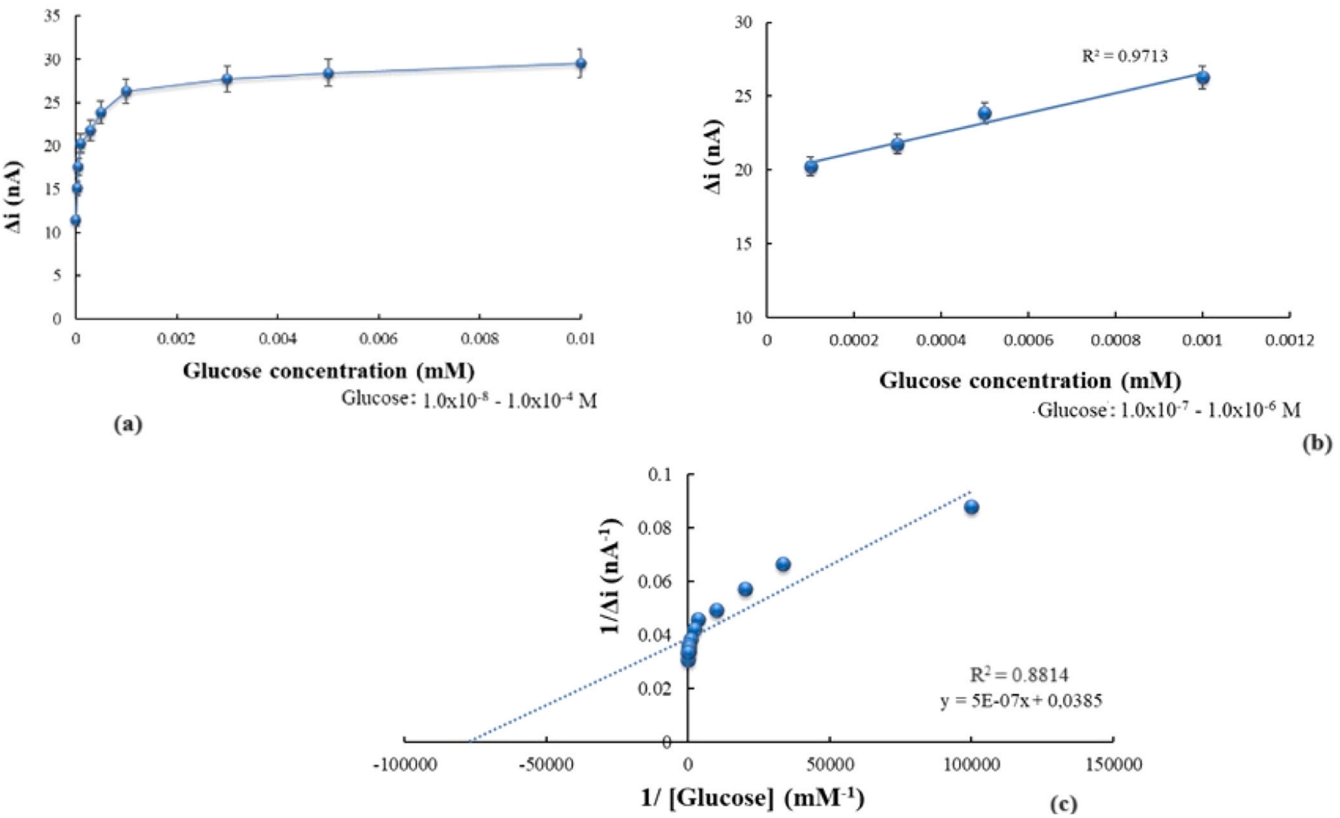

Fig. 10. (a) Effect of glucose concentration on the response of biosensor (MichaelMentencurve), (b) Calibration curve, (d) Lineweaver Burk curve (+0.4 V operating potential, at 25 °C, in 0.1 M pH 7.0 phosphate buffer) $R^2 = 0.8814$.

prepared biosensor, pH 5.0 (acetic acid sodium acetate buffer), pH 6.0, 7.0 and 8.0 (phosphate buffer (Na_2HP0_4 - NaH_2PO_4)) and pH 9.0 (glycine buffer) solutions were used separately. Glucose was added to cell (1.0×10^{-4} M) and current changes were calculated and this was repeated for all pH values. The prepared biosensor had the best activity at pH 7.0 (Fig. 9a). Although there are different pH values in glucose biosensor studies in the literature, there were generally pH values compatible with this study. For example; pH 8.5 (Tashkhourian et al., 2011), pH 7 (Aydoğdu et al., 2013), pH 8 (Özbek et al., 2021), pH 7.2 (Yang et al., 2009) and pH 7 (Wu et al., 2004). The reason for the difference in pH values could be related to the differences in used immobilization material and technique (Bodur et al., 2023).

Temperature is another important key factor for enzyme activity because of the degradation of enzymes at high temperatures. For this reason, the effect of the temperature on the designed biosensor performance must be identified.

To determine effects of temperature on designed biosensor, the cell temperature was settled to 20 °C with a constant flow using a thermostatic circulating water bath. Glucose (1.0×10^{-4} M) was added to the cell and the current changes were calculated at the end of the reaction. The same procedure was performed for 30, 40, 50, and 60 °C separately and the prepared biosensor had the best activity at 50 °C (Fig. 9b). There is glucose biosensor designed with different nanoparticles that had maximum activity at different temperatures in literature such as 25 ºC (Aydoğdu et al., 2013), 34.8 ºC (Yang et al., 2006), and 45 ºC (Wu et al., 2004). For the designed biosensor to be practical, the analyses were performed at room temperature.

3.5. Determination of substrate concentration effects

To determine the effect of substrate concentration on the prepared biosensor, the biosensor was equilibrated at +0.40 V potential in pH 7.0 buffer solution at 25 °C. Then, to obtain the Michaelis-Menten curve Fig. 10a), glucose solutions (1.0×10^{-8} - 1.0×10^{-3} M) were added to the cell, and current differences (Δi) were graphed against glucose concentration (Fig. 10b). Lineweaver–Burk curve (Fig. 10c) was graphed to calculate the K_m constant by graphing the data as 1/[Glucose]− 1/Δi. K_m value informs about the relationship between enzyme's catalytic activity and affinity of enzyme and substrate. A low K_m value indicates a high affinity and kinetic activity between enzyme and substrate. The $K_{m\ (app)}$ value was calculated as 0.0129 μM, and the I_{max} value was calculated as 38.5 μA. $K_{m(app)}$ values for immobilized glucose oxidase given in the literature are 18.0, 11.9, and 9.34 mM (Xue et al., 2001; Shan et al., 2009; Arslan et al., 2011). As a result of used silver nanoparticles, the designed biosensor has a high affinity for glucose with its low $K_{m(app)}$ value. The limit of detection (LOD) for glucose was evaluated as 5.0 nM with the formula LOD = $3\,s\,m^{-1}$ (where s is standard deviation of measurements and m is slope of the curve) and 200 s was determined as the response time and the linear working range was found as 0.10–1.0 μM. Linear range and detection limit in the literature is 0.1–8.0 mM (Cano et al., 2008), 0.2–150.0 mM (Kausaite- Minkstimiene et al., 2020) 10 μM - 0.8 mM (Wang et al., 2013); 0.1–2.5 mM (Ren et al., 2005), respectively. The detection limit is low when compared with the literature. Table 2 presents comparison of key analytical parameters with other glucose biosensors.

3.6. Determination of operational stability and storage stabilization of the glucose biosensor

The effect of electrode reusability was determined after successive measurements at a constant substrate concentration of 1.0×10^{-6} M in same day with same conditions. Δi values were plotted according to the number of measuring (Fig. 11a). The designed biosensor lost 9.19% of its initial activity after 12 measurements. The relative standard deviation was calculated as 3.97%.

To determine the shelf life of designed glucose biosensor, glucose

Table 2
Comparison of analytical characteristics of the different glucose biosensors.

Immobilization matrix	Electrochemical method and working potential	Immobilization techniques	Linearity	LOD	Km	Optimum pH	Optimum temperature	Long-term stability	Reference
Poly(1.10-phenanthrene-5.6-dione).poly(pyrrole-2-carboxylic acid)/ gold nanoparticles	Amperometric (+0.2 V)	Covalent bonding	0.2–150.0 mM	80 µM	-	4.0–9.0	-	14 days 96%	Kausaite-Minkstimiene et al., 2020
Silver nanoparticles and chitosan	Amperometric and cyclic voltammetry (-0.15 V)	More than one techniques	10 µM-15 mM	0.6977 µM	-	6.0–8.5	25^0C	9 days 93% 15 days 70%	Yang et al., 2014
Silver nanofibers and chitosan	Amperometrik ve siklik voltametri (-0.15 V)	More than one techniques	10 µM - 0.8 mM	2.83 µM	-	7.5	-	10 days 83%	Wang et al., 2013
Silver nanoparticles	Amperometric (+0.4 V)	Crosslinking	0.1–2.5 mM	-	-	-	-	-	Ren et al., 2005
Nafion	Amperometric (+0.84 V)	More than one techniques	0.02–15.0 mM	2.9 µM	-	-	-	-	Izadyar et al., 2021
Silica/lignin. Single-walled nanotubes	Voltammetry/ chronoamperometry (+0.3 V)	Adsorption	0.5–9 mM	145 µM	62 mM	7.0	25^0C	15 days 82% 21 days 73%	Jedrzak et al., 2018
Polypyrrole–polyvinyl sulfonate (Ppy–PVS) composite film	Amperometric (+ 0.7 V)	Crosslinking	-	-	6.25 mM	7.0	-	-	Gade et al., 2006
Polyaniline nafion graphene oxide	Amperometric (+0.3 V)	More than one techniques	0.5–50 mM	-	12.0 mM	6.0	-	-	Popov et al., 2021
WT- AgNPs modified CPE	**Amperometric (+0.4 V)**	**Crosslinking**	**5.0 nM**	**0.016 µM**	**0.0129 µM**	**7.0**	**50^0C**	**30 days 35%**	**This study**

(1.0×10^{-6} M) detection by using prepared biosensor was performed in 1., 3., 10., 15. and 25. days (Fig. 11 b). The decrease rate of enzyme activity was less from the 1st day to the 15th day. It was clear that the prepared biosensor kept 68.1% of its initial activity at the end of 15 days.

3.7. *Determination interfere effects*

The effects of ascorbic acid, which can be found in fruit juice and may cause interference in glucose detection were investigated. The ascorbic acid concentration was chosen as 1.0×10^{-4} M (Arslan et al., 2011). The designed biosensor was equilibrated at the optimum conditions, and the current changes (Δi) was recorded for 1.0×10^{-6} M glucose. Then, the ascorbic acid was added to the cell in a 10-fold dilution and the current change was measured. The interference effect of ascorbic acid was calculated using the obtained current changes. It was clear that ascorbic acid had an interference effect of 1.09%. The biosensor designed in this respect contributes sensitive and selective glucose determination.

3.8. *Glucose determination in fruit juice sample*

To determine the feasibility of the biosensor on real samples, the amount of glucose was determined by using the designed biosensor and compared with literature data. The commercial fruit juice solution was diluted 5.0×10^{5} times. The current change was measured and the glucose concentration in this fruit juice was calculated as 44.37 ± 4.14 g/L using the calibration curve. The analyses were repeated 3 times. The glucose amount in the same commercial fruit juice was determined by using LC/MS and was presented as 45.50 g/L (Walker et al., 2014). When the results were compared, it was clear that 99.98% accurate results were obtained with the designed biosensor.

4. Conclusion

Recycling the bioactive components contained in natural domestic waste, which increases due to the increasing population, and using them into the field of technology is very important in terms of sustainability. In this study, the bioactive components contained in the waste leaves consisting of the most consumed black tea were recovered and used in silver nanoparticle synthesis. The obtained nanoparticles were characterized and used in the design of an amperometric glucose biosensor, and the glucose amount of the commercial fruit juice sample was determined. The optimum operating potential of the designed biosensor was determined as +0.4 V and the response time was 200 s. The linear working range of the biosensor was determined as 0.10–1.0 µM and the LOQ was 0.016 µM. It has been determined that the biosensor designed by using nanoparticles obtained by recycling the bioactive components contained in waste tea leaves could determine the amount of glucose at very low concentrations. The glucose biosensor was found to have a suitable response time, wide operating range, low detection limit, good repeatability and long shelf life. Thus, it was clear that the bioactive compounds could be recycled and used in technology field.

CRediT authorship contribution statement

Merve Keskin: Writing – review & editing, Methodology, Data curation, Conceptualization. **Onur Can Bodur:** Writing – original draft, Formal analysis. **Fatma Arslan:** Writing – review & editing, Supervision, Project administration, Methodology, Data curation, Conceptualization. **Sevda Üçdemir Pektaş:** Writing – original draft, Formal analysis.

Declaration of Competing Interest

The authors confirm that they have no conflicts of interest with respect to the work described in this manuscript.

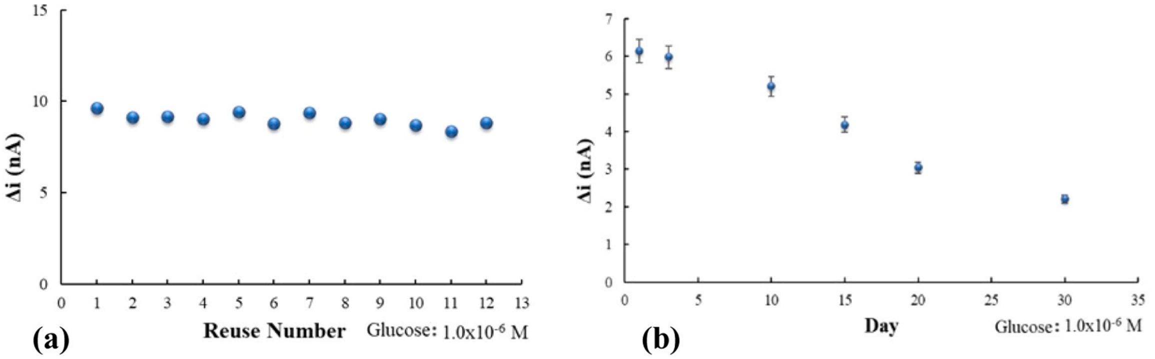

Fig. 11. Operational (a) and (b) storage stability of the biosensor (+0.4 V operating potential, at 25 °C, in 0.1 M pH 7.0 phosphate buffer).

Data availability

Data will be made available on request.

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